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Isotope shift in a beryllium atom

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We calculate the isotope shift of $2\,{}^1P_0 - 2\,{}^1S_0$, $3\,{}^1S_0 - 2\,{}^1S_0$ transitions and of the $2\,{}^1S_0$ ionization potential in the four-electron beryllium atom. The achieved precision is high enough to make possible the accurate determination of the nuclear charge radii from the experimental isotope shifts. This calls for corresponding measurements and opens the window for the determination of charge radii of heavier nuclei.

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I. INTRODUCTION

The determination of accurate charge radii from isotope shifts in atomic transitions was first performed for hydrogenic [1,2], and later for He [3–5], Li [6–8], and Be$^+$, systems [9–11]. While for hydrogenic ions the nonrelativistic wave function is known exactly, for all other systems it has to be obtained numerically, most often with the help of the variational approach. Presently, nonrelativistic energies of the He atom are known to more than 20 digits of accuracy [12–14]; for Li, it is about 15 digits [15,16]; and, very recently, the precision achieved for the Be atom reached 11 significant digits [17,18]. The computational approach for four-electron systems is based on explicitly correlated Gaussian (ECG) functions [19,20] and global optimization of more than 40 000 nonlinear parameters. All of the corrections to energies, including finite nuclear mass, relativistic and QED, are calculated as an expectation value with this nonrelativistic wave function.

The isotope shift observed in atomic spectra results from differences in the masses and charge distributions of the nuclei. For light isotopes, the mass shift is about 4–5 orders of magnitude larger than the shift caused by the finite size of the nucleus (the so-called field shift). Since it is not possible to separate the two effects experimentally, one has to rely on the theoretical calculations of the mass shift to extract the field shift effect from precise atomic spectroscopy. In this way, one determines the nuclear charge radius with respect to a well-known stable isotope, for which the charge radius is known from the electron-scattering experiments.

In this work, we perform the calculation of mass shift effects in transition energies of Be atoms with a precision more than sufficient to determine the nuclear charge radius from the experimental isotope shifts, once they become available. It will enable verification of the results obtained from the spectroscopy of the Be$^+$ ion, and also can be directly extended to other four-electron systems, e.g., the boron cation B$^+$ or doubly ionized carbon C^{2+}.

The most general approach to the calculation of energy levels of light atomic systems is based on the expansion in the fine-structure constant α. The leading term is the nonrelativistic energy. The higher-order coefficients in α, namely, the relativistic $m\alpha^4$ and quantum-electrodynamics (QED) $m\alpha^5$ corrections, are expressed as mean values of some effective Hamiltonians, while electron correlations are treated accurately by the use of explicitly correlated basis sets. For three-electron systems, the most accurate solution of the Schrödinger equation is obtained with the Hylleraas basis functions [15,16,21]. In such systems, the accuracy of the theoretical predictions for transition energies and isotope shifts is limited by the approximate treatment of higher-order ($m\alpha^6$ and $m\alpha^7$) QED corrections, rather than by numerical inaccuracies of the nonrelativistic wave function. Methods with Hylleraas functions have been extended to four-electron atomic systems, but only for some restricted selection of basis functions because of the significant difficulties in the evaluation of fully correlated integrals [22,23]. Even more difficult integrals appear in the matrix elements of relativistic operators. Unquestionably, significant efforts have to be made to improve upon the Hylleraas approach in order for it to be practical for four-electron systems. Therefore, at present, the method of choice for such systems is that based on explicitly correlated Gaussian (ECG) functions. The effectiveness of the ECG functions in treating few-electron problems has been demonstrated by many high-precision calculations of the nonrelativistic energies of atomic and molecular systems [20,24,25]. For the beryllium atom, the highest accuracy has been obtained using ECG functions [18,19,26–28]. In contrast to methods based on the Hylleraas functions, the main advantage of the ECG method is that the underlying integrations are manageable and very fast in numerical evaluations due to the compact formulas involving elementary functions only. On the other hand, the Gaussian functions have the drawback of improper asymptotic behavior since they decay too fast at long interparticle distances. They also have an incorrect short-range form and fail to correctly describe the Kato cusp. However, the two flaws can be overcome if one employs a sufficiently large and well-optimized ECG basis set. The issue is subtler in calculations of relativistic and QED properties, where the local inaccuracies of the wave functions result in significant numerical uncertainties of mean values. One has to very carefully optimize over a huge number of the variational parameters matching local behavior of the exact wave function as accurately as possible and employ dedicated techniques that accelerate the convergence of singular operators [29]. It is a time-consuming process, but the continuously ongoing advancements in accessible computing power and the development of new algorithms dedicated to multithreaded and parallel architecture make the extensive optimization of the Gaussian exponents computationally feasible even for systems with more than four electrons [20].

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II. ENERGY-LEVEL EXPANSION IN THE FINE-STRUCTURE CONSTANT

The energy level is considered as a function of the fine-structure constant α being expanded in a power series,

$$E(\alpha) = \sum_n E^{(n)}, \quad E^{(n)} \sim m\alpha^n, \quad n = 2,4,5,6,\ldots. \tag{1}$$

We perform also an expansion in the reduced electron mass to nuclear mass ratio $\eta = -\mu/M(\sim m/M)$,

$$E^{(n)} = \sum_k E^{(n,k)}, \quad E^{(n,k)} \sim m\alpha^n\eta^k \tag{2}$$

($k = 0,1,2,\ldots$), and each coefficient $E^{(n,k)}$ is calculated separately from an expectation value of the corresponding effective Hamiltonian.

The leading order $E^{(2)}$ is obtained from the nonrelativistic Hamiltonian in the center-of-mass system,

$$H^{(2)} = \sum_a \frac{p_a^2}{2m} + \frac{p_N^2}{2M} - \sum_a \frac{Z\alpha}{r_a} + \sum_{a<b} \frac{\alpha}{r_{ab}}, \tag{3}$$

with $\vec{p}_N = -\sum_a \vec{p}_a$. In order to extract a dependence on the finite nuclear mass M, all momenta and distances are scaled following the rules

$$\vec{p} \to \vec{p}\,(1+\eta), \quad \vec{r} \to \frac{1}{1+\eta}\vec{r}. \tag{4}$$

Then, the transformed nonrelativistic Hamiltonian (3) is given by

$$H^{(2)} = (1+\eta)\Bigg(H_0 - \eta\sum_{a<b}\frac{\vec{p}_a\cdot\vec{p}_b}{m}\Bigg). \tag{5}$$

The leading term $E^{(2,0)} \equiv E_0$ is a solution of the Schrödinger equation $H_0\Psi = E_0\Psi$ with the clamped nucleus Hamiltonian,

$$H_0 = \sum_a\left(\frac{p_a^2}{2m} - \frac{Z\alpha}{r_a}\right) + \sum_{a<b}\frac{\alpha}{r_{ab}}. \tag{6}$$

The mass polarization effect in Eq. (5) is included perturbatively. It leads to the first-order correction to the wave function $\tilde{\Psi} = \Psi + \eta\,\delta\Psi_{\rm mp}$ with

$$\delta\Psi_{\rm mp} = -\frac{1}{E_0 - H_0}\sum_{a<b}\frac{\vec{p}_a\cdot\vec{p}_b}{m}\,\Psi\,. \tag{7}$$

Let us introduce the following shorthand notation $\langle\ldots\rangle = \langle\Psi|\ldots|\Psi\rangle$ and $\langle\ldots\rangle_{\rm mp} = 2\,\langle\Psi|\ldots|\delta\Psi_{\rm mp}\rangle$. The first- and second-order nonrelativistic recoil coefficients can be written as

$$E^{(2,1)} = \eta\left(E^{(2,0)} - \sum_{a<b}\left\langle\frac{\vec{p}_a\cdot\vec{p}_b}{m}\right\rangle\right), \tag{8}$$

$$E^{(2,2)} = -\eta^2\sum_{a<b}\left(\left\langle\frac{\vec{p}_a\cdot\vec{p}_b}{m}\right\rangle + \frac{1}{2}\left\langle\frac{\vec{p}_a\cdot\vec{p}_b}{m}\right\rangle_{\rm mp}\right). \tag{9}$$

In calculations of relativistic effects for singlet states, the spin-spin interaction and the spin-orbit coupling terms vanish in the effective Breit-Pauli Hamiltonian [30]. We also do not consider the hyperfine structure. The final relativistic Hamiltonian is of the form

$$H^{(4)} = \sum_a H_a^{(4)} + \sum_{a<b} H_{ab}^{(4)} + \sum_a H_{aN}^{(4)}, \tag{10}$$

$$H_a^{(4)} = -\frac{\vec{p}_a^{\,4}}{8\,m^3}, \tag{11}$$

$$H_{ab}^{(4)} = \alpha\left\{\frac{\pi}{m^2}\,\delta^3(r_{ab}) - \frac{1}{2\,m^2}\,p_a^i\left(\frac{\delta^{ij}}{r_{ab}} + \frac{r_{ab}^i\,r_{ab}^j}{r_{ab}^3}\right)p_b^j\right\},$$

$$H_{aN}^{(4)} = Z\,\alpha\left\{\frac{1}{2\,m\,M}\,p_a^i\left(\frac{\delta^{ij}}{r_a} + \frac{r_a^i\,r_a^j}{r_a^3}\right)p_N^j + \frac{2\,\pi}{3}\left(\langle r_{\rm ch}^2\rangle + \frac{3}{4\,m^2}\right)\delta^3(r_a)\right\}. \tag{12}$$

All terms in Eq. (10) are treated as perturbations. Elements free of the nuclear mass and the rms radius $\langle r_{\rm ch}^2\rangle$ contribute to $E^{(4,0)}$ as mean values with the unperturbed wave function Ψ. Coefficient $E^{(4,1)}$ comprises terms proportional to η resulting from the scaling from Eq. (4) and the evaluation of the expectation value of $H^{(4)}$ with the perturbed wave function $\tilde{\Psi}$. Relativistic finite nuclear mass terms of order $O(m\alpha^4\eta^2)$ have been abandoned as negligible.

Analogously, the leading QED corrections $E^{(5,0)}$, $E^{(5,1)}$ are determined based on the known formulas [31,32]

$$E^{(5)} = -\frac{4\,Z\,\alpha^2}{3}\left(\frac{1}{m} + \frac{Z}{M}\right)^2\left\langle\sum_a\delta^3(r_a)\right\rangle\ln k_0 + \sum_a\langle H_{aN}^{(5)}\rangle + \sum_{a<b}\langle H_{ab}^{(5)}\rangle + E_{\rm pol}, \tag{13}$$

$$H_{aN}^{(5)} = \left[\frac{19}{30} + \ln(\alpha^{-2})\right]\frac{4\,\alpha^2\,Z}{3\,m^2}\,\delta^3(r_a) + \left[\frac{62}{3} + \ln(\alpha^{-2})\right]\frac{(Z\,\alpha)^2}{3\,m\,M}\,\delta^3(r_a) - \frac{7}{6\pi}\,\frac{m^2}{M}\,(Z\,\alpha)^5\,P\left(\frac{1}{(m\,\alpha\,r_a)^3}\right), \tag{14}$$

$$H_{ab}^{(5)} = \frac{\alpha^2}{m^2}\left(\frac{164}{15} + \frac{14}{3}\,\ln\alpha\right)\delta^3(r_{ab}) - \frac{7}{6\pi}m\alpha^5 P\left[\frac{1}{(m\alpha\,r_{ab})^3}\right], \tag{15}$$

where the Bethe logarithm and the Araki-Sucher distribution are defined by

$$\ln k_0 = \frac{\sum_{a,b}\left\langle\vec{p}_a\,(H_0 - E_0)\,\ln\left[\frac{2\,(H_0-E_0)}{\alpha^2\,m}\right]\vec{p}_b\right\rangle}{2\,\pi\,\alpha\,Z\,\sum_c\langle\delta^3(r_{\rm ch})\rangle}, \tag{16}$$

$$\langle\phi|P\left(\frac{1}{r^3}\right)|\psi\rangle = \lim_{a\to 0}\int d^3r\,\phi^*(\vec{r})\left[\frac{1}{r^3}\Theta(r-a) + 4\pi\,\delta^3(r)(\gamma + \ln a)\right]\psi(\vec{r}). \tag{17}$$

At present, complete numerical evaluation of the $m\alpha^6$ corrections for a four-electron system is unfeasible. Full calculations of the $E^{(6)}$ have been performed only for one- [33] and two-electron systems [34]. From this experience, we observe that the $E^{(6)}$ factor can be reasonably estimated using its dominating contribution built of one-electron terms,

$$E^{(6)} \approx \left\{ \frac{Z^2\,\alpha^3}{m^2}\left[\frac{427}{96} - 2\ln(2)\right] + \frac{Z^2\,\alpha^3}{m\,M}\left[\frac{35}{36} - \frac{448}{27\pi^2} - 2\ln(2) + \frac{6\zeta(3)}{\pi^2}\right] + \frac{Z^3\,\alpha^3}{m\,M}\left[4\ln(2) - \frac{7}{2}\right]\right\}\pi\sum_a \langle\delta^3(r_a)\rangle. \tag{18}$$

The above formula consists of electron-nucleus one-loop radiative, radiative recoil, and pure (no-loop) recoil corrections [33]. The electron-electron radiative corrections and the purely relativistic $m\alpha^6$ corrections were neglected. As in [18], the approximate treatment of $E^{(6)}$ is the main source of uncertainty in the overall error budget.

The nuclear recoil corrections $m\alpha^6\eta$ represented by coefficient $E^{(6,1)}$ are obtained from Eq. (18) in a procedure analogous to that described above for $E^{(4,1)}$ and $E^{(5,1)}$. Although the radiative recoil effect is negligible in the case of the transition energy calculations, it is still significant for the isotope mass shift. Again, the approximate form employed to estimate this contribution introduces some uncertainty in the determination of the isotope shift.

In addition to the above corrections resulting from the energy expansion (1) and (2), we included a term corresponding to the nuclear polarizability correction $E_{\rm pol}$. It comes from excitation of the nucleus by electrons orbiting in the atom. The relevant formula [7] relating the electric dipole nuclear transition moment with the shift of atomic-energy levels reads

$$E_{\rm pol} = -m\alpha^4\sum_a\langle\delta^3(r_a)\rangle(m^3\,\tilde{\alpha}_{\rm pol}), \tag{19}$$

where $\tilde{\alpha}_{\rm pol}$ is a *weighted* electric polarizability of the nucleus [7]. This correction has been calculated from the so-called $B(E1)$ function for beryllium isotopes ^{9,11}Be$^+$ [11,35]. In contrast to the beryllium ion calculations [11], we use in Eq. (19) the expectation value of the Dirac-δ operator of the neutral beryllium atom.

Following the convention introduced for $E^{(n,m)}$, formulas for related contributions to the transition energy and to the isotope mass shift are given by

$$\nu^{(n,m)}(X \to Y) = E^{(n,m)}(X) - E^{(n,m)}(Y), \tag{20}$$

$$\Delta\nu^{(n,m)}(A,B) = \nu^{(n,m)}(A) - \nu^{(n,m)}(B). \tag{21}$$

One of the main goals of this work was to present data which enable determination of the nuclear charge radius of one isotope, say A, with respect to B,

$$\delta r^2_{\rm ch} = \langle r^2_{\rm ch}\rangle_A - \langle r^2_{\rm ch}\rangle_B. \tag{22}$$

$\delta r^2_{\rm ch}$ can be determined from the difference between the experimental and theoretical isotope mass shifts,

$$\Delta\nu^{\rm exp} - \Delta\nu^{\rm the}_{\rm ms} = C\,\delta r^2_{\rm ch}, \tag{23}$$

provided that the C constant is known. To determine this constant, we consider various corrections due to the finite nuclear size. The leading $r_{\rm ch}$-dependent corrections consist of the $m\alpha^4$ terms extracted from Eq. (12),

$$E^{(4,0)}_{\rm fs} = \frac{2\pi}{3}Z\alpha\langle r^2_{\rm ch}\rangle\sum_a\langle\delta^3(r_a)\rangle \tag{24}$$

and

$$E^{(4,1)}_{\rm fs} = \frac{2\pi}{3}Z\alpha\langle r^2_{\rm ch}\rangle\eta\left[3\sum_a\langle\delta^3(r_a)\rangle + \sum_a\langle\delta^3(r_a)\rangle_{\rm mp}\right]. \tag{25}$$

We include also the logarithmic relativistic correction to the wave function at the origin,

$$E^{(6,0)}_{\rm fs,log} = -(Z\,\alpha)^2\ln\left(Z\alpha m\langle r^2_{\rm ch}\rangle\right)E^{(4,0)}_{\rm fs}. \tag{26}$$

Our recommended value of the constant C is obtained as a sum of three components, $C = C^{(4,0)} + C^{(4,1)} + C^{(6,0)}_{\rm log}$.

III. NUMERICAL CALCULATIONS

In numerical calculations, we use dimensionless energies $\mathcal{E}^{(n,k)}$ with the α and η factors pulled out, e.g., $E^{(n,k)} = m\alpha^n\eta^k\mathcal{E}^{(n,k)}$. The same can also be done for all terms in the effective Hamiltonian $H^{(n,k)} = m\alpha^n\eta^k\mathcal{H}^{(n,k)}$ entertaining transformations $p \to m\alpha p$ and $r^{-1} \to m\alpha r^{-1}$.

As an extension of our previous nonrelativistic results for $2\,^1S$ and $2\,^1P$ states [18], we present herein results for the 3^1S state. We employed the ECG basis functions of progressively doubled size from 512 to 4096 terms. The sequence of energies obtained for several basis sets enables estimation of the final energy error. The energy obtained from the largest basis is of comparable accuracy to the value published a few years ago by Stanke *et al.* [27], but the size of our basis set has been significantly reduced thanks to the thorough optimization.

Apart from finding the best possible energy and the wave function of the unperturbed atom, one of the most crucial parts of our perturbative calculations of the isotope mass shift is an accurate representation of the mass polarization correction to the wave function, $\delta\Psi_{\rm mp}$, of Eq. (7). This correction can be found as a solution of the inhomogeneous equation

$$(\mathcal{E}_0 - \mathcal{H}_0)\delta\Psi_{\rm mp} = -\sum_{a<b}(\vec{p}_a\cdot\vec{p}_b - \langle\vec{p}_a\cdot\vec{p}_b\rangle)\Psi. \tag{27}$$

The operator $\vec{p}_a\cdot\vec{p}_b$ changes neither the orbital angular momentum nor the spin symmetry of Ψ. This means that $\delta\Psi_{\rm mp}$ can be expressed using basis functions of the same symmetry as Ψ. The variational parameters of such a basis can be determined in a minimization of the symmetric quantity $\mathcal{J}[\delta\Psi_{\rm mp}] = \langle\sum_{a<b}\vec{p}_a\cdot\vec{p}_b\rangle_{\rm mp}$. In our approach, the basis set for $\delta\Psi_{\rm mp}$ is divided into two sectors. The first sector is built of the known basis functions with the nonlinear parameters determined in the minimization of $\mathcal{E}_0$. For this purpose, we selected one of the previously generated basis sets of size

equal to half of the final size of Ψ. The nonlinear parameters of this basis remain fixed during the optimization in order to enable accurate representation of the states orthogonal to Ψ. The second sector, of size equal to that of the final Ψ, consists of basis functions that undergo optimization with respect to the functional $\mathcal{J}$. Due to the more complicated structure of the first-order function, both the convergence and the cost of the optimization are less favorable than in the case of the unperturbed wave function optimization. The first-order correction function $\delta\Psi_{\rm mp}$ obtained in the procedure sketched above is subsequently employed to evaluate all the mass polarization corrections for the relativistic and QED operators.

Direct use of formulas (10) and (13) for relativistic and QED operators leads to a slow numerical convergence of its mean values, since they are sensitive to local inaccuracies of the wave function Ψ and the mass polarization correction $\delta\Psi_{\rm mp}$. This spurious effect is especially observable with the ECG functions having improper short-distance behavior. The solution is to employ the regularized matrix elements following Drachman‘s recipes [36]. Pertinent expressions for the relativistic operators, including those with the mass polarization correction, have been presented in the Appendix. Below we provide the set of formulas for the Araki-Sucher distribution $P(r^{-3})$. The expression for $P(r_{ab}^{-3})$ has been derived in Ref. [29],

$$\big[P\big(r_a^{-3}\big)\big]_r = 4\pi(1+\gamma)\delta^3(r_a) + 4(\mathcal{E}_0-\mathcal{V})r_a^{-1}\ln r_a - 2\sum_b \vec{p}_b r_a^{-1}\ln r_a\,\vec{p}_b, \tag{28}$$

$$\big[P\big(r_{ab}^{-3}\big)\big]_r = 4\pi(1+\gamma)\delta^3(r_{ab}) + 2(\mathcal{E}_0-\mathcal{V})r_{ab}^{-1}\ln r_{ab} - \sum_c \vec{p}_c\, r_{ab}^{-1}\ln r_{ab}\,\vec{p}_c, \tag{29}$$

where $\mathcal{V}$ is a dimensionless Coulomb potential. Such a regularized operator, denoted as $[\ldots]_r$, has exactly the same expectation value as the operator inside the square brackets if the exact wave function is used.

In the regularized form of the second-order elements $\langle\ldots\rangle_{\rm mp}$, there are additional first-order terms to be evaluated following the formula

$$\big\langle P\big(r_{ab}^{-3}\big)\big\rangle_{\rm mp} = \big\langle\big[P\big(r_{ab}^{-3}\big)\big]_r\big\rangle_{\rm mp} + 2\sum_{c<d}\big(\big\langle\vec{p}_c\cdot\vec{p}_d\, r_{ab}^{-1}\ln r_{ab}\big\rangle - \langle\vec{p}_c\cdot\vec{p}_d\rangle\big\langle r_{ab}^{-1}\ln r_{ab}\big\rangle\big). \tag{30}$$

The Bethe logarithm and its mass polarization correction is computationally the most demanding term in this work. It involves numerical evaluation of the integrand in 200 points in the method based on the integral representation introduced by Schwartz [37,38]. Recently, we have obtained the Bethe logarithms for the $2\,^1S$ and $2\,^1P$ states [18], whereas here we present a result for the $3\,^1S$ state. The method of evaluation of the mass polarization correction to the Bethe logarithm was originally applied to the helium atom [39]. We have recently extended this approach to the lithium atom [8], and we follow this approach here.

IV. RESULTS

Numerical results for all operators necessary to evaluate the transition energies and the isotope shifts with the $2\,^1S$, $3\,^1S$, and $2\,^1P$ states of the beryllium atom are presented in Table I. In calculations of the isotope mass shift of the ionization energy, we use the numerical values obtained previously with the Hylleraas basis functions [11] for the Be$^+$ ion. These values appear to be determined with much higher numerical precision than that accessible from the ECG approach. In Table I, some of the first-order quantities $\langle\ldots\rangle$ for the $2\,^1S$ and $2\,^1P$ states come from our former work [18]. For a given operator, the final value is assessed in an extrapolation from a series of basis sets of progressively doubled size. We have used up to 4096 basis functions to represent the $2\,^1S$ and $3\,^1S$ states in the first-order expectation value as well as in the mass polarization corrections $\langle\ldots\rangle_{\rm mp}$. The exception is the Bethe logarithm calculated with, at most, 2048 basis functions. For the $2\,^1P$ state, the most accurate results have been acquired from a 6144-term basis, but only for the first-order terms. The expressions for the expectation values in the ^1P states are much more complex compared to those of the ^1S states, and thus accurate determination of the mass polarization corrections is very time consuming. We obtained these corrections with

TABLE I. Dimensionless mean values for $2\,^1S$, $3\,^1S$, and $2\,^1P$ states of the beryllium atom.

Operator	$2\,^1S$		$3\,^1S$		$2\,^1P$	
	$\langle\ldots\rangle$	$\langle\ldots\rangle_{\rm mp}$	$\langle\ldots\rangle$	$\langle\ldots\rangle_{\rm mp}$	$\langle\ldots\rangle$	$\langle\ldots\rangle_{\rm mp}$
$\mathcal{H}_0$	−14.667 356 498(3)		−14.418 240 37(5)		−14.473 451 37(4)	
$\vec{p}_a\cdot\vec{p}_b$	0.460 224 112(8)	7.702 088 9(11)	0.450 500 94(9)	6.930 24(12)	0.434 811 25(13)	8.291 95(5)
$\vec{p}_a^{\,4}$	2 165.630 1(9)	138.347(6)	2 148.339 7(18)	122.58(18)	2 133.321 1(12)	195.83(11)
$\delta^3(r_a)$	35.369 002 6(6)	1.274 91(17)	35.127 977(9)	1.039(5)	34.897 914 6(8)	2.268(3)
$\delta^3(r_{ab})$	1.605 305 33(9)	−0.417 38(9)	1.583 185 4(7)	−0.425 2(6)	1.567 943 6(2)	−0.335 7(5)
$p_a^i\big(\frac{\delta^{ij}}{r_{ab}}+\frac{r_{ab}^i r_{ab}^j}{r_{ab}^3}\big)\,p_b^j$	1.783 648 19(15)	19.320 040(3)	1.800 258 3(15)	18.243 7(5)	1.624 185 8(5)	19.278 7(8)
$p_N^i\big(\frac{\delta^{ij}}{r_a}+\frac{r_a^i r_a^j}{r_a^3}\big)\,p_a^j$	224.965 525(6)		222.843 53(7)		220.689 8(2)	
$P\big(r_{ab}^{-3}\big)$	−7.326 766(3)	−0.305(7)	−7.472 06(4)	−0.12(5)	−7.097 15(8)	−1.978(9)
$P\big(r_a^{-3}\big)$	−917.750 9(2)		−911.495 2(9)		−905.256(3)	
$\ln(k_0)$	5.750 46(2)	−0.130 9(2)	5.751 49(5)	−0.126 7(7)	5.752 32(8)	−0.128 8(11)

TABLE II. Dimensionless coefficients of the energy difference expansion in powers of α and η. For the $3\,{}^1S-2\,{}^1S$ transition, the coefficients were explicitly converted to MHz using CODATA [41] inverse fine-structure constant $\alpha^{-1}=137.035\,999\,074(44)$ and the nuclear mass $m_N(^9\text{Be})=9.012\,182\,20(43)$ u [42]. The finite-size correction $\nu^{(4,0)}_{\text{fs}}$ was calculated with $r_{\text{ch}}(^9\text{Be})=2.519(12)$ fm [43].

Contribution	$3\,{}^1S-2\,{}^1S$	(MHz)	$2\,{}^1P-2\,{}^1S$	IP($2\,{}^1S$)
$\nu^{(2,0)}$	0.249 116 13(6)	54 674.671(13)	0.193 905 149(8)	0.342 593 324(6)
$\nu^{(2,1)}$	0.258 839 3(2)	−3.458 8	0.219 318 0(2)	0.349 898 28(3)
$\nu^{(2,2)}$	0.395 65(6)		−0.269 541(12)	0.677 212(4)
$\nu^{(4,0)}$	0.569 5(8)	6.65 6(9)	1.041 2(2)	0.634 38(12)
$\nu^{(4,1)}$	0.632(8)		−2.060(3)	0.926 4(5)
$\nu^{(5,0)}$	−5.945(10)	−0.507(1)	−11.849(16)	−6.542(5)
$\nu^{(5,1)}$	−4.74(15)		28.86(22)	−7.36(5)
$\nu^{(6,0)}$	−37.(7)	−0.023(4)	−73.(15)	−41.(9)
$\nu^{(6,1)}$	−199.(100)		−165.(83)	−232.(116)
$\nu^{(4,0)}_{\text{fs}}$	$-2.019\,21(7)\langle r_c^2\rangle$	−0.001 0	$-3.946\,57(2)\langle r_c^2\rangle$	$-2.211\,23(1)\langle r_c^2\rangle$
Total		54 677.337(17)		
Theor. [27].		54 677.378(30)		
Expt. [40].		54 677.26(10)		

3072-term functions, and the Bethe logarithm calculations involved up to 1536 basis functions.

Table II contains dimensionless coefficients of the energy difference ensuing from the expansions (1) and (2), which have been computed on the basis of the data collected in Table I. These coefficients enable summation of the expansion for any isotope. We show explicitly the results of such a summation for the $3\,{}^1S-2\,{}^1S$ energy gap in the ^{9}Be isotope (see column 3 of Table II). A major contribution to the overall uncertainty of this value comes from the assumption that the missing $m\alpha^6$ and higher-order terms contribute approximately 20% of the evaluated correction $\nu^{(6,0)}$. For this transition, we have obtained agreement with the other theoretical calculation [27] as well as with the experimental value [40]. Note, however, that theoretical results are currently significantly more accurate than measurement results. The entries neglected in the third column of Table II correspond to contributions that do not affect the absolute transition frequencies. However, they are significant in the isotope shift calculations. The results of analogous summation for the $2\,{}^1P-2\,{}^1S$ transition and the $2\,{}^1S$ ionization potential have already been given in [18].

The expansion coefficients $\nu^{(n,1)}, n=2,4,5,6$ and $\nu^{(2,2)}$ of Table II can be used to evaluate subsequent contributions to the isotope mass shift $\Delta\nu^{\text{the}}_{\text{ms}}$. An example of such an evaluation for the ^{11}Be–^{9}Be shift is presented in Table III. We observe that the leading nonrelativistic contribution gives at least 99.9% of the total isotope shift. The relativistic and QED recoil corrections are small but still important. The uncertainty of our $\Delta\nu^{\text{the}}_{\text{ms}}$ comes in 50% from $\Delta\nu^{(6,1)}$, in 10% from $\Delta\nu_{\text{pol}}$, and in the remaining part from several smaller contributions. The nucleus polarizability $\tilde{\alpha}_{\text{pol}}$ has been determined based on $B(E1)$ data [11,35] and is used here to obtain the shift of energy levels E_{pol}. Calculating coefficient C, we included the finite mass correction as well as the logarithmic relativistic correction, resulting in a small isotope dependence. The uncertainty of C coefficients comes from the estimation of the relativistic correction to the wave function at the origin, which is about 25% of the logarithmic part. This completes the theoretical results necessary for the nuclear charge radii determination of the beryllium isotopes.

V. CONCLUSION

The principal motivation for this work was to provide theoretical means for determination of the nuclear radii from the isotope shifts of transition lines $2\,{}^1P-2\,{}^1S$, $3\,{}^1S-2\,{}^1S$, and ionization potential of the $2\,{}^1S$ state. The uncertainty of our calculations for beryllium isotope mass shifts is dominated by the numerical uncertainty of $\Delta\nu^{(n,1)}, n=2,4$, and the estimation of unknown terms of order $m\alpha^6\eta$, which amounts

TABLE III. Contributions to the ^{11}Be–^{9}Be isotope shift of the $3\,{}^1S-2\,{}^1S$, $2\,{}^1P-2\,{}^1S$ transition and ionization potential IP($2\,{}^1S$) in MHz, with exclusion of the finite-size correction. The second uncertainty of $\Delta\nu^{\text{the}}_{\text{ms}}$ is due to the atomic mass. The nucleus polarizability $\tilde{\alpha}_{\text{pol}}(^{11}\text{Be})=6.90(69)\times10^{-7}$ m^{-3} [11], $\tilde{\alpha}_{\text{pol}}(^9\text{Be})=2.90(29)\times10^{-7}$ m^{-3} [35].

Contribution	$3\,{}^1S-2\,{}^1S$	$2\,{}^1P-2\,{}^1S$	IP($2\,{}^1S$)
$\Delta\nu^{(2,1)}$	18 907.131(15)	16 020.271(15)	25 558.619(2)
$\Delta\nu^{(2,2)}$	−3.198 2(5)	2.178 8(1)	−5.474 2
$\Delta\nu^{(4,1)}$	2.46(3)	−8.013(12)	3.604(2)
$\Delta\nu^{(5,1)}$	−0.135(4)	0.819(6)	−0.209(2)
$\Delta\nu^{(6,1)}$	−0.041(20)	−0.034(17)	−0.048(24)
$\Delta\nu_{\text{pol}}$	0.034(3)	0.066(7)	0.037(4)
$\Delta\nu^{\text{the}}_{\text{ms}}$	18 906.25(4)(1)	16 015.29(3)(1)	25 556.53(3)(1)
C(MHz/fm^2)	−4.772(8)	−9.334(16)	−5.225(9)

to 30–40 kHz. This is sufficient to determine the mean-square charge radii difference with a precision of about 0.003 fm^2, which is much less than the uncertainty in the charge radius of the reference nucleus. The beryllium rms charge radii of $^{7,10\text{-}12}$Be have already been obtained relative to the only stable ^{9}Be nucleus from the D_1 and D_2 transition lines in three-electron Be$^+$ [9,44]. The results provided herein can be employed for a consistency check with corresponding values extracted from presumably more accurate transitions in atomic beryllium. More important, however, is that the analogous results can be obtained for the berylliumlike ions, such as the boron cation B$^+$ or carbon cation C^{2+}.

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APPENDIX: REGULARIZATION OF RELATIVISTIC OPERATORS

In this section, we present regularization formulas. The following three operators, denoted by $[\dots]_r$, are defined as

$$\sum_a [p_a^4]_r = 4\,(\mathcal{E}_0 - \mathcal{V})^2 - 2 \sum_{a<b} \vec{p}_a^{\,2}\, \vec{p}_b^{\,2}, \tag{A1}$$

$$4\pi\,[\delta^3(r_a)]_r = 4\,(\mathcal{E}_0 - \mathcal{V})\, r_a^{-1} - 2 \sum_b \vec{p}_b\, r_a^{-1}\, \vec{p}_b\,, \tag{A2}$$

$$4\pi\,[\delta^3(r_{ab})]_r = 2\,(\mathcal{E}_0 - \mathcal{V})\, r_{ab}^{-1} - \sum_c \vec{p}_c\, r_{ab}^{-1}\, \vec{p}_c. \tag{A3}$$

For the exact wave function, the following expectation value identities are valid:

$$\sum_a \langle p_a^4\rangle = \sum_a \langle [p_a^4]_r\rangle, \tag{A4}$$

$$\langle 4\pi\,\delta^3(r_a)\rangle = \langle 4\pi\,[\delta^3(r_a)]_r\rangle, \tag{A5}$$

$$\langle 4\pi\,\delta^3(r_{ab})\rangle = \langle 4\pi\,[\delta^3(r_{ab})]_r\rangle, \tag{A6}$$

$$\sum_a \langle p_a^4\rangle_{\mathrm{mp}} = \sum_a \langle [p_a^4]_r\rangle_{\mathrm{mp}} - 8 \sum_{a<b} (\langle \vec{p}_a \cdot \vec{p}_b\, \mathcal{V}\rangle - 2\,\mathcal{E}_0\, \langle \vec{p}_a \cdot \vec{p}_b\rangle), \tag{A7}$$

$$\langle 4\pi\,\delta^3(r_a)\rangle_{\mathrm{mp}} = \langle 4\pi\,[\delta^3(r_a)]_r\rangle_{\mathrm{mp}} + 4 \sum_{b<c} \big(\langle \vec{p}_b \cdot \vec{p}_c\, r_a^{-1}\rangle - \langle \vec{p}_b \cdot \vec{p}_c\rangle \langle r_a^{-1}\rangle\big), \tag{A8}$$

$$\langle 4\pi\,\delta^3(r_{ab})\rangle_{\mathrm{mp}} = \langle 4\pi\,[\delta^3(r_{ab})]_r\rangle_{\mathrm{mp}} + 2 \sum_{c<d} \big(\langle \vec{p}_c \cdot \vec{p}_d\, r_{ab}^{-1}\rangle - \langle \vec{p}_c \cdot \vec{p}_d\rangle \langle r_{ab}^{-1}\rangle\big). \tag{A9}$$

For an approximate wave function, the right-hand side converges faster to the exact value than does the conventional expression on the left-hand side.

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