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RAPID COMMUNICATION

Precision calculation of ^{4,6,8}He isotope shifts

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Standard perturbation theory is employed to calculate the mass shifts of the $2\,^1S_0$–$2\,^3S_1$ and $2\,^3S_1$–$2\,^3P_J$ transitions for ^{4,6,8}He. High-precision results are obtained for the mass shifts in the isotope pairs ^{6}He–^{4}He and ^{8}He–^{4}He, with uncertainties below 1 part per million (ppm). Our analysis provides a complete set of isotope-shift results and systematically examines their sensitivity to nuclear charge-radius differences. Once experimental measurements reach a precision comparable to that of the calculated mass shifts, the squared differences of nuclear charge radii can be determined with an accuracy of approximately 0.4%–0.6%, representing an order-of-magnitude improvement over current values.

Keywords: isotope shifts, nuclear charge radius, atomic spectroscopy, quantum electrodynamic

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1. Introduction

The accuracy of atomic spectroscopy has improved dramatically over the past few decades, driven by advances in experimental techniques. As a result, it has become a powerful tool for probing nuclear properties and searching for new physics beyond the Standard Model.[1–7] Its applications span a wide range of areas, including the precise determination of nuclear charge radii in helium isotopes,[4,8] the observation of atomic parity violation in ^{133}Cs,[9] the investigation of possible variations in fundamental constants using atomic clocks,[10,11] and tests of the weak equivalence principle via atom interferometry.[12] Moreover, recent studies suggest that linearity in King plots can serve as a sensitive probe of higher-order isotope shifts and constrain the coupling strength of hypothetical new bosonic fields to electrons and neutrons.[13–16]

Helium and helium-like ions represent the most fundamental few-electron atomic systems, enabling the application of well-established theoretical methods to perform highly accurate calculations of their electronic structures.[17–22] Of particular significance is the precise determination of nuclear charge-radius differences between helium isotopes through isotope-shift measurements, which has emerged as a major focus of contemporary research. This field has advanced rapidly, driven by a combination of high-precision experimental techniques and state-of-the-art theoretical calculations.[17,18,23,24]

Recently, Rengelink *et al.*[25] and Werf *et al.*[26] measured the $2\,^1$S–$2\,^3S$ transition in both ^{4}He and ^{3}He, yielding a refined value for the nuclear charge-radius difference of 1.0757(15) fm^2. Meanwhile, Schuhmann *et al.*[27] determined the nuclear charge-radius difference between ^{3}He and ^{4}He by measuring the 2S–2P transitions in muonic helium ions, μ^3He$^+$ and μ^4He$^+$, obtaining a value of 1.0636(31) fm^2. This result revealed a 3.6σ discrepancy when compared with the atomic transition data, prompting Schuhmann *et al.*[27] to speculate that the anomaly could hint at new physics beyond the Standard Model.

In response, Muli *et al.*[28] carried out refined calculations of nuclear-structure corrections in an attempt to resolve the inconsistency, but the discrepancy persisted. Our recent work addressed this issue by incorporating the previously neglected hyperfine-mixing effect in ^{3}He,[29] which was subsequently validated and improved upon by Pachucki *et al.*[30] This led to an updated value of 1.0678(7) fm^2 derived from the $2\,^1$S–$2\,^3$S transition, bringing it into agreement with the muonic helium result within 1.3σ.

More recently, Wen *et al.*[31] revisited their analysis of the $2\,^3$S–$2\,^3$P transition in ^{4}He, accounting for post-selection shifts, and updated the nuclear charge-radius difference to 1.0733(21) fm^2. This revision has reintroduced a 2.6σ discrepancy relative to the muonic helium determination, under-

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scoring the ongoing challenges and complexity involved in resolving this fundamental problem.

As an isotope of ^{4}He, the ^{6}He nucleus is formed by adding two neutrons to a ^{4}He core. It is stable under the strong interaction and has a relatively long half-life of approximately 0.8 s. Due to the inherent stability and strong binding of the ^{4}He core, the ^{6}He nucleus can be modeled as a three-body system consisting of an alpha particle and two neutrons. Remarkably, the removal of any one of the three components leads to an unbound system, a feature that characterizes ^{6}He as a "Borromean nucleus".[32] Another intriguing isotope, ^{8}He, is a neutron-rich nucleus with the highest known neutron-to-proton ratio of 3 among bound light nuclei. Its structure consists of a ^{4}He core surrounded by four valence neutrons and has a half-life of about 0.1 s. In ^{8}He, the four neutrons are loosely bound and spatially extended, forming a diffuse neutron halo.

In 2013, Lu *et al.*[33] determined the nuclear charge radii of ^{6}He and ^{8}He to be 2.059(8) fm and 1.958(16) fm, respectively, by measuring isotope shifts in the $2\,^3\mathrm{S}_1$–$3\,^3\mathrm{P}_\mathrm{J}$ transitions. Owing to their exotic structures and rich dynamics, ^{6}He and ^{8}He serve as valuable systems for probing fundamental nuclear forces. Detailed studies of their nuclear structures provide important benchmarks for developing effective nuclear-interaction models capable of accurately describing both binding mechanisms and internal correlations.[34,35]

In the present work, we calculate the isotope shifts of the $2\,^1\mathrm{S}_0$–$2\,^3\mathrm{S}_1$ and $2\,^3\mathrm{S}_1$–$2\,^3\mathrm{P}_\mathrm{J}$ transitions for the ^{6}He–^{4}He and ^{8}He–^{4}He pairs, including recoil corrections up to order $m\alpha^6$. The theoretical framework is presented in Section 2, where all relevant corrections — nonrelativistic recoil, relativistic recoil, quantum electrodynamic (QED) recoil, and nuclear polarizability — are discussed in detail. The numerical results and their analysis are provided in Section 3, followed by a summary in Section 4. Atomic units (a.u.) are used throughout unless otherwise specified.

2. Theoretical method

The isotope shift E_{IS} consists of two components, the mass shift E_{MS} and the field shift E_{FS},

$$E_{\mathrm{IS}} = E_{\mathrm{MS}} + E_{\mathrm{FS}}, \tag{1}$$

where E_{IS} is obtained from experimental measurements, while E_{MS} must be calculated with high theoretical precision. The field shift E_{FS} contains information about the nuclear charge-radius difference ΔR^2. Neglecting nuclear-polarization effects, it can be expressed as

$$E_{\mathrm{FS}} = C\Delta R^2, \tag{2}$$

where $C = C_X - C_Y$, and C_X is the field-shift coefficient for state X, given to lowest order by

$$C_X = \frac{2\pi Ze^2}{3}\left\langle \sum_i \delta(\boldsymbol{r}_i)\right\rangle_X. \tag{3}$$

The nonrelativistic quantum electrodynamics (NRQED) framework is employed to compute the mass shift E_{MS}. Within NRQED, atomic energy levels are expanded in powers of the fine-structure constant α as

$$E(\alpha) = E^{(2)} + E^{(4)} + E^{(5)} + E^{(6)} + E^{(7)} + \cdots, \tag{4}$$

where each term is defined as $E^{(n)} \equiv m\alpha^n\varepsilon^{(n)}$, and

$$\varepsilon^{(n)} = \varepsilon_0^{(n)} + \varepsilon_1^{(n)} + \varepsilon_2^{(n)} + \cdots, \tag{5}$$

with $\varepsilon_0^{(n)}$, $\varepsilon_1^{(n)}$, and $\varepsilon_2^{(n)}$ denoting the zeroth-, first-, and second-order recoil corrections in the electron-to-nucleus mass ratio m/M, respectively. Here, m and M are the masses of the electron and the nucleus.

In this work, we employ the Hylleraas variational method to determine high-precision wave functions corresponding to the nonrelativistic Hamiltonian in the infinite-nuclear-mass limit

$$H_0 = \frac{\boldsymbol{p}_1^2}{2} + \frac{\boldsymbol{p}_2^2}{2} - \frac{Z}{r_1} - \frac{Z}{r_2} + \frac{1}{r}, \tag{6}$$

where $\boldsymbol{r} = \boldsymbol{r}_1 - \boldsymbol{r}_2$ and Z is the nuclear charge. The Hylleraas basis set[36] employed in the variational expansion is given by

$$\psi_{ijk}(\boldsymbol{r}_1, \boldsymbol{r}_2) = r_1^i r_2^j r^k\, \mathrm{e}^{-\alpha r_1 - \beta r_2 - \gamma r}\mathrm{Y}_{l_1 l_2}^{LM}(\hat{r}_1, \hat{r}_2), \tag{7}$$

where $\mathrm{Y}_{l_1 l_2}^{LM}(\hat{r}_1, \hat{r}_2)$ denotes the vector-coupled product of spherical harmonics associated with the angular momenta of the two electrons. Based on this wave function, higher-order corrections arising from relativistic and QED effects — specifically those associated with the finite nuclear mass — are subsequently evaluated order by order. The explicit expressions for the correction operators included at each order of the calculation are provided as follows.

2.1. Nonrelativistic recoil

The nonrelativistic Hamiltonian $H_M^{(2)}$ for two-electron atomic (ionic) systems is represented as a sum of two terms

$$H_M^{(2)} = \mu H_\infty^{(2)} + \frac{\mu^2}{m} H_{\mathrm{rec}}^{(2)}, \tag{8}$$

where $H_\infty^{(2)} \equiv H_0$ is the nonrelativistic Hamiltonian of the system in the center-of-mass frame, $\mu = \frac{mM}{m+M}$ is the reduced mass, and

$$H_{\mathrm{rec}}^{(2)} = \frac{m}{M}\boldsymbol{p}_1 \cdot \boldsymbol{p}_2 \tag{9}$$

is the mass-polarization term. Since the nuclear mass M is much larger than the electron mass m, the effect of this recoil

Hamiltonian $H^{(2)}_{\rm rec}$ is treated perturbatively. The normal mass shift arises from $\mu H^{(2)}_{\infty}$, which has the following form:

$$E^{(2)}_{M,0} = (\gamma-1)\langle H^{(2)}_{\infty}\rangle, \tag{10}$$

where $\gamma \equiv \mu/m$. The second recoil correction is induced by $H^{(2)}_{\rm rec}$, which is known as the specific mass shift or mass polarization. In principle, all orders of the m/M corrections in the nonrelativistic case can be obtained by computing the eigenvalue difference of the nonrelativistic Hamiltonian with and without the recoil term $H^{(2)}_{\rm rec}$. However, this approach does not allow for a clear separation of contributions from individual orders in m/M. Therefore, we evaluate the mass-polarization corrections using standard perturbation theory, including terms up to third order. These perturbation corrections are

$$E^{(2)}_{M,1} = \gamma^2\langle H^{(2)}_{\rm rec}\rangle, \tag{11}$$

$$E^{(2)}_{M,2} = \gamma^3\langle H^{(2)}_{\rm rec}, H^{(2)}_{\rm rec}\rangle, \tag{12}$$

$$E^{(2)}_{M,3} = \gamma^4[\langle H^{(2)}_{\rm rec}, H^{(2)}_{\rm rec}, H^{(2)}_{\rm rec}\rangle - \langle H^{(2)}_{\rm rec}\rangle\langle H^{(2)}_{\rm rec}, H^{(2)}_{\rm rec}\rangle_2], \tag{13}$$

where we define

$$\langle A, B\rangle \equiv \left\langle A \frac{1}{(E_0-H_0)'} B\right\rangle, \tag{14}$$

$$\langle A, B\rangle_2 \equiv \left\langle A \frac{1}{(E_0-H_0)'^2} B\right\rangle, \tag{15}$$

$$\langle A, B, C\rangle \equiv \left\langle A \frac{1}{(E_0-H_0)'} B \frac{1}{(E_0-H_0)'} C\right\rangle, \tag{16}$$

with E_0 denoting the eigenvalue of the nonrelativistic Hamiltonian in the nonrecoil limit.

2.2. Relativistic recoil

The expectation value of the Breit–Pauli Hamiltonian $H^{(4)}$ represents the relativistic correction of order $m\alpha^4$ to the nonrelativistic energy. The Breit–Pauli Hamiltonian including the full reduced-mass dependence is[23]

$$\begin{aligned} H^{(4)} = \gamma^3\Bigg[& -\gamma\frac{p_1^4+p_2^4}{8} + \frac{Z\pi}{2}\left(\delta^3(r_1)+\delta^3(r_2)\right) - \frac{1}{2}p_1^i\left(\frac{\delta^{ij}}{r} + \frac{r^i r^j}{r^3}\right)p_2^j - (\boldsymbol{S}^2 - \boldsymbol{S}_{\rm A}^2)\frac{2\pi}{3}\delta^3(r) \\ & -\pi\delta^3(r) + \frac{1}{4}(S^iS^j - S^i_{\rm A}S^j_{\rm A})\left(\frac{\delta^{ij}}{r^3} - 3\frac{r^i r^j}{r^5}\right) + \frac{3}{4}\boldsymbol{S}\cdot\frac{\boldsymbol{r}}{r^3}\times(\boldsymbol{p}_2-\boldsymbol{p}_1) + \frac{1}{4}\boldsymbol{S}_{\rm A}\cdot\frac{\boldsymbol{r}}{r^3}\times(\boldsymbol{p}_2+\boldsymbol{p}_1) \\ & +\frac{Z}{4}\boldsymbol{S}\cdot\left(\frac{\boldsymbol{r}_1}{r_1^3}\times\boldsymbol{p}_1 + \frac{\boldsymbol{r}_2}{r_2^3}\times\boldsymbol{p}_2\right) + \frac{Z}{4}\boldsymbol{S}_{\rm A}\cdot\left(\frac{\boldsymbol{r}_1}{r_1^3}\times\boldsymbol{p}_1 - \frac{\boldsymbol{r}_2}{r_2^3}\times\boldsymbol{p}_2\right)\Bigg], \end{aligned} \tag{17}$$

where $\boldsymbol{S} = (\boldsymbol{\sigma}_1+\boldsymbol{\sigma}_2)/2$ and $\boldsymbol{S}_{\rm A} = (\boldsymbol{\sigma}_1-\boldsymbol{\sigma}_2)/2$ denote the total and antisymmetric spin operators corresponding to the triplet and singlet states, respectively.

The recoil correction arising from the reduced-mass dependence in Eq. (17) is

$$\varepsilon^{(4)}_{M,0} = \langle H^{(4)}\rangle - \langle H^{(4)}_{\infty}\rangle. \tag{18}$$

The first-order recoil correction, $\varepsilon^{(4)}_{M,1}$, consists of two parts: (i) the recoil contribution to the Breit Hamiltonian and (ii) the second-order perturbative correction involving the nonrecoil Breit Hamiltonian and the mass-polarization operator. It can be expressed as

$$\varepsilon^{(4)}_{M,1} = \gamma^3\langle H^{(4)}_{\rm rec}\rangle + 2\gamma\langle H^{(4)}_{\infty}, H^{(2)}_{\rm rec}\rangle, \tag{19}$$

where $H^{(4)}_{\rm rec}$ is the recoil correction to $H^{(4)}$, defined by

$$\begin{aligned} H^{(4)}_{\rm rec} = \frac{Zm}{2M}\Bigg[& \left(\frac{\boldsymbol{r}_1}{r_1^3} + \frac{\boldsymbol{r}_2}{r_2^3}\right)\times\boldsymbol{P}\cdot\boldsymbol{S} + \left(\frac{\boldsymbol{r}_1}{r_1^3} - \frac{\boldsymbol{r}_2}{r_2^3}\right)\times\boldsymbol{P}\cdot\boldsymbol{S}_{\rm A} \\ & -p_1^i\left(\frac{\delta^{ij}}{r_1} + \frac{r_1^i r_1^j}{r_1^3}\right)P^j - p_2^i\left(\frac{\delta^{ij}}{r_2} + \frac{r_2^i r_2^j}{r_2^3}\right)P^j\Bigg], \end{aligned} \tag{20}$$

where $\boldsymbol{P} \equiv \boldsymbol{p}_1 + \boldsymbol{p}_2$ is the total electron momentum operator.

The second-order recoil correction can be decomposed into two contributions

$$\varepsilon^{(4)}_{M,2} = \varepsilon^{(4)}_{M,2a} + \varepsilon^{(4)}_{M,2b}, \tag{21}$$

where

$$\begin{aligned} \varepsilon^{(4)}_{M,2a} = {} & 2\langle H^{(4)}_{\infty}, H^{(2)}_{\rm rec}, H^{(2)}_{\rm rec}\rangle + \langle H^{(2)}_{\rm rec}, H^{(4)}_{\infty}, H^{(2)}_{\rm rec}\rangle \\ & -\langle H^{(4)}_{\infty}\rangle\langle H^{(2)}_{\rm rec}, H^{(2)}_{\rm rec}\rangle_2 - 2\langle H^{(2)}_{\rm rec}\rangle\langle H^{(4)}_{\infty}, H^{(2)}_{\rm rec}\rangle_2, \end{aligned} \tag{22}$$

$$\varepsilon^{(4)}_{M,2b} = 2\langle H^{(4)}_{\rm rec}, H^{(2)}_{\rm rec}\rangle. \tag{23}$$

The first part, $\varepsilon^{(4)}_{M,2a}$, represents the third-order perturbation of $H^{(4)}$ with two mass-polarization operators $H^{(2)}_{\rm rec}$, whereas the second part, $\varepsilon^{(4)}_{M,2b}$, corresponds to the second-order perturbation involving $H^{(4)}_{\rm rec}$ and $H^{(2)}_{\rm rec}$.

Higher-order relativistic effects mainly originate from the second-order perturbation of $H^{(4)}$ and the first-order perturbation of the effective Hamiltonian of order $m\alpha^6$. The calculation of these contributions, with the relevant formulas provided in Refs. [17,18,37], is highly challenging. Nevertheless, their overall impact is small, and their effects are estimated and included as part of the final uncertainty.

2.3. QED recoil

In the nonrecoil limit, the leading QED correction $H^{(5)}_{\infty}$ is given by

$$H^{(5)}_{\infty} = \frac{4Z}{3}\left(\ln(Z\alpha)^{-2} + \frac{19}{30} - \ln\frac{k_0}{Z^2}\right)\left(\delta^3(r_1)+\delta^3(r_2)\right)$$

$$+\left(\frac{14}{3}\ln(Z\alpha)+\frac{164}{15}\right)\delta^3(r)$$
$$-\frac{14}{3}\left(\frac{1}{4\pi r^3}+\delta^3(r)\ln Z\right), \tag{24}$$

where $\ln k_0$ is the Bethe logarithm, as defined in Ref. [23]. The expectation value of the singular operator $1/r^3$ is obtained through a renormalization procedure, as described in Ref. [38].

The recoil correction consists of three parts

$$\varepsilon^{(5)}_{M,\mathrm{QED}}=\frac{m}{M}\left(\varepsilon^{(5)}_{M,1}+\varepsilon^{(5)}_{M,2}\right)+\varepsilon^{(5)}_{M,3}, \tag{25}$$

where

$$\varepsilon^{(5)}_{M,1}=-3\langle H^{(5)}_{\infty}\rangle+\frac{4Z}{3}\langle\delta^3(r_1)+\delta^3(r_2)\rangle-\frac{14}{3}\langle\delta^3(r)\rangle, \tag{26}$$

$$\varepsilon^{(5)}_{M,2}=Z^2\left(-\frac{2}{3}\ln(Z\alpha)+\frac{62}{9}-\frac{8}{3}\ln k_0\right)\langle\delta^3(r_1)+\delta^3(r_2)\rangle$$
$$-\frac{7Z^2}{6\pi}\left\langle\frac{1}{r_1^3}+\frac{1}{r_2^3}\right\rangle, \tag{27}$$

$$\varepsilon^{(5)}_{M,3}=\langle H^{(5)}_{\infty},H^{(2)}_{\mathrm{rec}}\rangle-\frac{4Z}{3}\ln\left(\frac{k_0'}{Z^2}\right)\langle\delta^3(r_1)+\delta^3(r_2)\rangle. \tag{28}$$

The $\ln k_0'$ term represents the first-order perturbation of the Bethe logarithm due to the mass-polarization operator $H^{(2)}_{\mathrm{rec}}$.

The higher-order QED recoil correction is of order $m^2\alpha^6/M$ and can be expressed as

$$\varepsilon^{(6)}_{M,\mathrm{QED}}=\varepsilon^{(6)}_{M,1}+\varepsilon^{(6)}_{M,2}+\varepsilon^{(6)}_{M,3}+\varepsilon^{(6)}_{M,4}, \tag{29}$$

where

$$\varepsilon^{(6)}_{M,1}=(\gamma^3-1)\left[Z^2\pi\left(\frac{427}{96}-2\ln 2\right)\langle\delta^3(r_1)+\delta^3(r_2)\rangle+\pi\left(\frac{6}{\pi^2}\zeta(3)-\frac{697}{27\pi^2}-8\ln 2+\frac{1099}{72}\right)\langle\delta^3(r)\rangle\right], \tag{30}$$

$$\varepsilon^{(6)}_{M,2}=(\gamma^3-1)\left[Z\pi\left(-\frac{2179}{648\pi^2}-\frac{10}{27}+\frac{3}{2}\ln 2-\frac{9}{4\pi^2}\zeta(3)\right)\langle\delta^3(r_1)+\delta^3(r_2)\rangle\right.$$
$$\left.+\pi\left(\frac{15}{2\pi^2}\zeta(3)+\frac{631}{54\pi^2}-5\ln 2+\frac{29}{27}\right)\langle\delta^3(r)\rangle\right], \tag{31}$$

$$\varepsilon^{(6)}_{M,3}=Z^2\frac{m}{M}\left(\frac{35}{36}-\frac{448}{27\pi^2}-2\ln 2+\frac{6}{\pi^2}\zeta(3)\right)\langle\delta^3(r_1)+\delta^3(r_2)\rangle, \tag{32}$$

$$\varepsilon^{(6)}_{M,4}=Z^3\frac{m}{M}\left(4\ln 2-\frac{7}{2}\right)\langle\delta^3(r_1)+\delta^3(r_2)\rangle. \tag{33}$$

These four terms originate from three distinct sources: the reduced-mass scaling of the one-loop QED contribution, the radiative recoil contribution, and the pure recoil contribution.

2.4. Nuclear polarizability

The nuclear polarizability correction to the energy levels can be expressed as

$$\varepsilon^{(4)}_{\mathrm{pol}}=-\widetilde{\alpha}_{\mathrm{pol}}\cdot\langle\delta^3(r_1)+\delta^3(r_2)\rangle, \tag{34}$$

where the coefficient $\widetilde{\alpha}_{\mathrm{pol}}$ has been evaluated as $\widetilde{\alpha}_{\mathrm{pol}}(^4\mathrm{He})=2.07(20)$ fm^3 and $\widetilde{\alpha}_{\mathrm{pol}}(^6\mathrm{He})=24.7(5.0)$ fm^3.[33,39] Slightly smaller values for ^{4}He were reported in Ref. [40], but the differences are not significant at the level of accuracy relevant to the present work. The correction is negligibly small for ^{8}He.

3. Results and discussion

The main objective of this section is to numerically analyze the mass shifts, field shifts, and isotope shifts of the $2\,^1\mathrm{S}$–$2\,^3\mathrm{S}$ and $2\,^3\mathrm{S}$–$2\,^3\mathrm{P}$ transitions for the ^{6}He–^{4}He and ^{8}He–^{4}He isotope pairs. Here, the Bethe logarithms $\ln(k_0/Z^2)$ are adopted from Ref. [37], with values of 2.9801183651(1) for the $2\,^1\mathrm{S}_0$ state, 2.97774245929(2) for the $2\,^3\mathrm{S}_1$ state, and 2.9836910033(2) for the $2\,^3\mathrm{P}_\mathrm{J}$ states. The first-order perturbative corrections to the Bethe logarithm arising from the mass polarization operator, denoted as $\frac{M}{m}\ln(k_0)_M$, are 0.141875(1) for $2\,^1\mathrm{S}_0$, 0.0382843(1) for $2\,^3\mathrm{S}_1$, and 0.069676(1) for $2\,^3\mathrm{P}_\mathrm{J}$.

Table 1. Numerical results for the DK operators of the $2\,^3\mathrm{P}_\mathrm{J}$ states, in atomic units.

Operator	^{4}He	^{6}He	^{8}He
$H^1_{\mathrm{DK}}:\nabla_1^2(\boldsymbol{r}_1\times\boldsymbol{p}_1)/r_1^3$	−0.36626544	−0.36622131	−0.36619933
$H^2_{\mathrm{DK}}:(\boldsymbol{r}_1\times\boldsymbol{r})(\boldsymbol{r}\cdot\boldsymbol{p}_2)/r^3r_1^3$	−0.01004596	−0.01004858	−0.01004989
$H^3_{\mathrm{DK}}:r^ir_1^j/r^3r_1^3$	0.00227147	0.00227483	0.00227650
$H^4_{\mathrm{DK}}:(\boldsymbol{r}\times\boldsymbol{p}_2)/r^4$	−0.08464419	−0.08464545	−0.08464608
$H^5_{\mathrm{DK}}:r^ir^j/r^6$	−0.03629056	−0.03629187	−0.03629252
$H^6_{\mathrm{DK}}:\nabla_1^2(\boldsymbol{r}\times\boldsymbol{p}_1)/r^3$	−0.18242308	−0.18241730	−0.18241444
$H^7_{\mathrm{DK}}:\nabla_1^2(\boldsymbol{r}\times\boldsymbol{p}_2)/r^3$	0.27952588	0.27955530	0.27956998
$H^8_{\mathrm{DK}}:\nabla_1^2(\boldsymbol{p}_1\times\boldsymbol{p}_2)/r$	0.00111834	0.00110617	0.00110011
$H^9_{\mathrm{DK}}:\nabla_1^2(\boldsymbol{r}\cdot\boldsymbol{p}_2)(\boldsymbol{r}\times\boldsymbol{p}_1)/r^3$	−0.01050638	−0.01051852	−0.01052453
$H^{10}_{\mathrm{DK}}:[\boldsymbol{r}\times(\boldsymbol{r}\cdot\boldsymbol{p}_2)\boldsymbol{p}_1]/r^5$	−0.02831713	−0.02831831	−0.02831891
$H^{11}_{\mathrm{DK}}:\{\boldsymbol{r}\times(\boldsymbol{r}\times\boldsymbol{p}_1)^i\boldsymbol{p}_2\}^j/r^5$	0.00531108	0.00531253	0.00531326
$H^{12}_{\mathrm{DK}}:\boldsymbol{p}_2^i\boldsymbol{p}_1^j/r^3$	−0.03694155	−0.03694152	−0.03694152
$H^{13}_{\mathrm{DK}}:\nabla_1^2r^ir^j/r^5$	0.28701288	0.28704167	0.28705603
$H^{14}_{\mathrm{DK}}:\nabla_1^2r^ip_1^j/r^3$	0.12135504	0.12136460	0.12136940
$H^{15}_{\mathrm{DK}}:\nabla_1^2r^ip_2^j/r^3$	0.04763768	0.04764227	0.04764452

The numerical results for the various operators presented above are taken from Ref. [29] and its supplementary material. Tables 1 and 2 list the numerical values of the Douglas–Kroll (DK) operators and their recoil corrections, with uncertainties smaller than one unit in the last significant digit.

Table 2. Numerical results for the recoil corrections to the DK operators of the $2\,^3\mathrm{P}_\mathrm{J}$ states, in atomic units.

Operator	He
$V_1 : p_1^2(\boldsymbol{p}_1\times\boldsymbol{p}_2)/r_1$	−0.918584
$V_2 : p_1^2\boldsymbol{r}_1\cdot[(\boldsymbol{r}_1\times\boldsymbol{p}_1)^i(\boldsymbol{p}_1+\boldsymbol{p}_2)]/r_1^3$	0.316347
$V_3 : p_1^2\boldsymbol{r}_1\times(\boldsymbol{p}_1+\boldsymbol{p}_2)/r_1^3$	−0.704255
$V_4 : \boldsymbol{r}\times(\boldsymbol{p}_1+\boldsymbol{p}_2)/r_1r^3$	−0.061736
$V_5 : \boldsymbol{r}\times\boldsymbol{r}_1[\boldsymbol{r}_1\cdot(\boldsymbol{p}_1+\boldsymbol{p}_2)]/r_1^3r^3$	−0.020237
$V_6 : \boldsymbol{r}_1\times\boldsymbol{r}_2(\boldsymbol{r}_1\cdot\boldsymbol{p}_1)/r_1^3r_2^3$	0.056545
$V_7 : \boldsymbol{r}_1\times(\boldsymbol{p}_1+\boldsymbol{p}_2)/r_1^4$	−0.225347
$V_8 : r_2^ir_1^j/r_2^3r_1^3$	0.061829

Table 3 presents the isotope shifts for the $2\,^1\mathrm{S}$–$2\,^3\mathrm{S}$ and $2\,^3\mathrm{S}$–$2\,^3\mathrm{P}$ transitions in the ^{6}He–^{4}He and ^{8}He–^{4}He isotope pairs. The first four rows list the nonrelativistic nuclear recoil corrections of order $m\alpha^2$, as given by Eqs. (10)–(13). The following three rows contain the relativistic recoil corrections of order $m\alpha^4$, computed using Eqs. (18), (19), and (21). The eighth row provides the QED recoil correction of order $m\alpha^5$, derived from Eq. (25). The subsequent row includes higher-order relativistic and QED contributions of order $m\alpha^6$, which are estimated using the approximation in Eq. (29) due to the complexity of a full calculation. The dominant source of uncertainty in the results arises from the omission of terms of order $m\alpha^6$ and higher. These uncertainties constitute a major contribution to the total error in the computed mass shifts. The row labeled "NPol" represents the nuclear polarization correction, as given by Eq. (34). The final row reports the total mass shifts obtained by summing all the above contributions. These values are determined with high precision, and the associated uncertainties are estimated to be below 1 ppm.

Also in the table, for the field shift, the following coefficients C in Eq. (2) are used:

$$C(2\,^1\mathrm{S}\text{–}2\,^3\mathrm{S}) = -214.45\ \mathrm{kHz/fm^2}, \tag{35}$$

$$C(2\,^3\mathrm{S}\text{–}2\,^3\mathrm{P}) = 1210.5\ \mathrm{kHz/fm^2}. \tag{36}$$

These values can be compared with previously reported results[23] $C(2\,^1\mathrm{S}\text{–}2\,^3\mathrm{S}) = -214.66(2)$ kHz/fm^2, $C(2\,^1\mathrm{S}\text{–}2\,^3\mathrm{S}) = -214.40$ kHz/fm^2, $C(2\,^3\mathrm{S}\text{–}2\,^3\mathrm{P}) = 1212.2(1)$ kHz/fm^2, and $C(2\,^3\mathrm{S}\text{–}2\,^3\mathrm{P}) = 1209.8$ kHz/fm^2. The differences in squared nuclear charge radii are $\Delta\langle r^2\rangle = 1.415(7)$ fm^2 for the ^{6}He–^{4}He pair, and $1.009(42)$ fm^2 for the ^{8}He–^{4}He pair.[33]

Table 3 indicates that the dominant source of uncertainty in the field shifts arises from the error in the differences of nuclear charge radii, which also constrains the overall precision of the isotope shift measurements. By referencing Eq. (1), the relationship between the uncertainty in the squared nuclear charge radii difference and the errors in the isotope and mass shifts can be expressed as

$$\delta\Delta R^2 = \frac{\sqrt{\delta E_{\mathrm{IS}}^2 + \delta E_{\mathrm{MS}}^2}}{C}.$$

If the precision of the experimentally measured isotope shift is comparable to that of the mass shift, i.e., $|\delta E_{\mathrm{IS}}| \approx |\delta E_{\mathrm{MS}}|$ (since $E_{\mathrm{IS}} \approx E_{\mathrm{MS}}$), the relative uncertainty in ΔR^2 can be estimated by

$$\zeta = \left|\frac{\delta\Delta R^2}{\Delta R^2}\right| = \sqrt{2}\left|\frac{\delta E_{\mathrm{MS}}}{E_{\mathrm{FS}}}\right|.$$

Table 3. Spin-independent contributions to the mass shifts, in kHz. When not explicitly stated, the numerical uncertainty is estimated to be less than 1 in the last significant digit.

	^{6}He–^{4}He		^{8}He–^{4}He	
	$2\,^1\mathrm{S}$–$2\,^3\mathrm{S}$	$2\,^3\mathrm{S}$–$2\,^3\mathrm{P}$	$2\,^1\mathrm{S}$–$2\,^3\mathrm{S}$	$2\,^3\mathrm{S}$–$2\,^3\mathrm{P}$
$\mu\alpha^2$	8839787.30	−12710411.5	13240877.87	−19038580.9
$\mu\alpha^2(\mu/M)$	−622831.01	−21754942.5	−932943.02	−32586884.0
$\mu\alpha^2(\mu/M)^2$	5363.03	−10165.5	7228.47	−13701.5
$\mu\alpha^2(\mu/M)^3$	−0.16	−2.4	−0.20	−3.0
$\mu\alpha^4$	9170.52	−18305.1	13736.78	−27420.0
$\mu\alpha^4(\mu/M)$	−6614.77	20568.3	−9908.65	30810.1
$\mu\alpha^4(\mu/M)^2$	−1.93	5.5	−2.60	7.4
$m\alpha^5(m/M)$	−57.94	62.1	−86.79	93.0
$m\alpha^6(m/M)$	−2.38(60)	13.5(3.4)	−3.57(94)	20.2(5.1)
NPol	3.00(60)	−16.9(3.4)	−0.27(3)	1.5(2)
E_{MS}	8224815.66(85)	−34473194.7(4.8)	12318898.01(94)	−51635657.2(5.1)
E_{FS}	−303.5(1.5)	1713.2(8.4)	−216.4(8.9)	1222(50)
E_{IS}	8224512.2(1.7)	−34471481.5(9.7)	12318681.6(9.0)	−51634436(51)
$\lvert E_{\mathrm{FS}}/E_{\mathrm{IS}}\rvert$	36.9 ppm	49.7 ppm	17.6 ppm	23.7 ppm
ζ	0.4 %	0.4 %	0.6 %	0.6 %

Table 4. Spin-dependent contributions to the mass shifts of the $2\,^3P_J$ states for the ^{6,8}He–^{4}He isotope pairs, in kHz. When not explicitly stated, the numerical uncertainty is estimated to be less than 1 in the last significant digit.

	^{6}He–^{4}He			^{8}He–^{4}He		
	$2\,^3P_0$	$2\,^3P_1$	$2\,^3P_2$	$2\,^3P_0$	$2\,^3P_1$	$2\,^3P_2$
$\mu\alpha^4$	−3797.0232	275.1460	594.3170	−5687.5883	412.1432	890.2317
$\mu\alpha^4(\mu/M)$	4110.1506	315.6675	−1011.4306	6156.5499	472.8631	−1515.0278
$\mu\alpha^4(\mu/M)^2$	2.0970	0.0927	−0.4750	2.8263	0.1250	−0.6402
$\mu\alpha^5(\mu/M)$	−4.0486	1.0063	0.2059	−6.0642	1.5073	0.3084
$\mu\alpha^6(\mu/M)$	0.057(57)	−0.085(85)	0.039(39)	0.088(88)	−0.127(127)	0.059(59)
Sum	311.23(6)	591.83(9)	−417.34(4)	465.81(9)	886.51(13)	−625.07(6)

Table 5. Spin-dependent contributions to the mass shifts of the $2\,^3P_J$ fine-structure splittings between ^{6,8}He and ^{4}He, i.e., the splitting isotope shifts (SIS), in kHz.

Isotope pair	$2\,^3P_1$–$2\,^3P_0$	$2\,^3P_2$–$2\,^3P_1$
^{6}He–^{4}He	280.24(11)	−1008.81(10)
^{8}He–^{4}He	420.16(16)	−1511.04(14)

Table 3 presents further details on the ratios $|E_{\rm FS}/E_{\rm IS}|$ and the corresponding values of ζ. When the experimental uncertainties in isotope shifts and mass shifts are comparable, the accuracy in determining the squared nuclear charge radii differences can be significantly enhanced, reaching a relative uncertainty level of approximately 0.4%–0.6%.

The spin-dependent contributions to the $2\,^3P_J$ states are summarized in Table 4, where the $m^2\alpha^6/M$-order correction is presented as an approximate value derived solely from the DK operators and their associated recoil corrections. A conservative uncertainty estimate of 100% has been assigned to this correction, as it constitutes the principal source of error in the overall calculation. A detailed numerical breakdown of the DK contributions and recoil terms is provided in Tables 1 and 2. Moreover, the effect of singlet-triplet mixing in the $2\,^3P_1$ state is considerably more significant than in the $2\,^3P_0$ and $2\,^3P_2$ states, resulting in shifts of the values in Table 4 from 591.83(9) kHz and 886.51(13) kHz to 591.47(9) kHz and 885.97(13) kHz. Considering this effect and the data in Table 4, we evaluate the isotope shifts of the fine-structure splittings, referred to as the splitting isotope shifts (SIS),[41,42] which are presented in Table 5. These SIS values play a crucial role in testing the internal consistency of experimental results.

4. Summary

In this work, we have calculated the nuclear mass corrections for the $2\,^1S$, $2\,^3S$, and $2\,^3P$ states of ^{4,6,8}He using perturbation theory. The mass shifts for the $2\,^1S$–$2\,^3S$ and $2\,^3S$–$2\,^3P$ transitions were computed with uncertainties below 1 ppm. The contributions and uncertainties of the field shifts were evaluated based on the currently available differences in nuclear charge radii. We find that the dominant source of error in the isotope shifts arises from the uncertainty in the field shifts, which amounts to approximately 15 ppm–50 ppm. Our calculations demonstrate that once experimental measurements achieve precision comparable to that of the theoretical mass shifts, the determination of nuclear charge radius differences can be improved to an accuracy of 0.4%–0.6%, thereby enabling stringent tests of nuclear structure models for neutron-rich helium isotopes.

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