Three-photon-exchange nuclear structure correction in hydrogenic systems

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The complete relativistic $O(\alpha^2)$ nuclear structure correction to the energy levels of ordinary (electronic) and muonic hydrogenlike atoms is investigated. The elastic part of the nuclear structure correction is derived analytically. The resulting formula is valid for an arbitrary hydrogenic system and is much simpler than analogous expressions previously reported in the literature. The analytical result is verified by high-precision numerical calculations. The inelastic $O(\alpha^2)$ nuclear structure correction is derived for the electronic and muonic deuterium atoms. The correction comes from a three-photon exchange between the nucleus and the bound lepton and has not been considered in the literature so far. We demonstrate that in the case of deuterium, the inelastic three-photon-exchange contribution is of a similar size and of the opposite sign to the corresponding elastic part and, moreover, cancels exactly the model dependence of the elastic part. The obtained results affect the determination of nuclear charge radii from the Lamb shift in ordinary and muonic atoms.

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I. INTRODUCTION

The determination of the nuclear charge radii from atomic spectra is a very interesting test of the Standard Model of fundamental interactions. The lepton universality, namely, the identical interaction strength of all leptons, ensures that the nuclear charge radii derived from the ordinary (electronic) and the muonic atoms should be exactly the same. However, a series of experiments on μH [1] and μD [2] and (still unpublished) measurements on $\mu\,^3$He and $\mu\,^4$He [3] revealed significant discrepancies for the determined nuclear charge radii, as compared to those derived from the corresponding electronic atoms. To verify these discrepancies one should carefully examine all possible sources of uncertainties in the spectroscopic determinations of the nuclear charge radii.

The main theoretical uncertainty of the Lamb shift in light muonic atoms comes from our insufficient knowledge of the nuclear internal structure. The nuclear structure corrections are usually divided into the elastic and the inelastic part. The elastic part (also referred to as the finite nuclear size correction) is induced by a static distribution of the nuclear charge and can be obtained by solving the Dirac equation. The inelastic nuclear correction is much more complicated; it encompass the nuclear dipole polarizability and higher-order contributions. To deal with the nuclear corrections, one performs an expansion of the binding energy in powers of the fine structure constant α and examines the expansion terms one after another.

The leading nuclear effect is of order α^4 and of pure elastic origin. The first-order $O(\alpha)$ nuclear-structure correction (often referred to as the two-photon exchange contribution) has both elastic and inelastic parts and was extensively studied both for the electronic and the muonic atoms [4–8]. One of the interesting results was a significant cancellation between the elastic and the inelastic $O(\alpha)$ nuclear contributions.

The next-order $O(\alpha^2)$ nuclear structure correction comes from the three-photon exchange between the bound lepton and the nucleus. Only the elastic part of this correction has been addressed in the literature so far [9]. In the present work we demonstrate that the inelastic $O(\alpha^2)$ contribution is significant and partially cancels its elastic counterpart. We also derive formulas for the complete $O(\alpha^2)$ nuclear correction in deuterium. Our calculation is performed in the nonrecoil limit and neglects the magnetic dipole and electric quadrupole moments of the nucleus. The results obtained affect determinations of nuclear charge radii from the precision spectroscopy of ordinary and muonic atoms. However, they are not able to explain the previously reported discrepancy between the H-D and μH-μD isotope shift [2].

We now introduce notations for the nuclear radii that will be extensively used throughout this paper. r_C denotes the root-mean-square (rms) charge radius of an arbitrary nucleus, $r_C \equiv \sqrt{\langle r^2\rangle}$. We will use specific notations for several important nuclei: $r_C(\mathrm{H}) \equiv r_p$ for the rms radius of the proton, $r_C(\mathrm{D}) \equiv r_d$ for the rms radius of the deuteron, and r_s for the deuteron structure radius $r_s^2 = r_d^2 - r_p^2$. Since we neglect the finite nuclear mass effects, there is no $3/(4\,m_p^2)$ term in r_d^2. We define $r_{CC} = \sqrt[4]{\langle r^4\rangle}$ for an arbitrary nucleus, with the specific cases of $r_{CC}(\mathrm{H}) \equiv r_{pp}$ for the proton, $r_{CC}(\mathrm{D}) \equiv r_{dd}$ for the

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deuteron, and r_{ss} for the corresponding structure radius of the deuteron. r_Z is the third Zemach moment defined below by Eq. (15). We will also introduce two new effective nuclear radii of arbitrary nuclei, r_{C1} and r_{C2}, defined by Eqs. (62) and (66), respectively. The corresponding specific notations are $r_{C1}(\mathrm{H}) \equiv r_{p1}$ and $r_{C2}(\mathrm{H}) \equiv r_{p2}$ for the proton and r_{d1} and r_{d2} for the deuteron, respectively.

II. LEADING FINITE NUCLEAR SIZE CORRECTION

In this section we rederive well-known results for the leading nuclear correction of order α^4, which is of pure elastic (finite nuclear size) origin and induced by the one-photon exchange between the bound lepton and the nucleus. This derivation sets the ground for our further evaluation of higher-order corrections.

Let us assume that the nucleus is a scalar particle with the charge density $\rho(q^2)$ in the momentum space. The electron-nucleus interaction potential in momentum space is then

$$V(q^2) = -\rho(q^2)\,\frac{4\pi\,Z\alpha}{q^2}. \tag{1}$$

The expansion coefficients of ρ in q^2,

$$\rho(q^2) = 1 + \rho'(0)\,q^2 + \tfrac{1}{2}\,\rho''(0)\,q^4 + \dots, \tag{2}$$

can be interpreted in terms of momenta of the nuclear charge distribution $\langle r^2\rangle$ and $\langle r^4\rangle$,

$$\rho'(0) = -\frac{\langle r^2\rangle}{6}, \tag{3}$$

$$\rho''(0) = \frac{\langle r^4\rangle}{60}. \tag{4}$$

From the second term in the right-hand side of Eq. (2), one immediately obtains the leading finite nuclear size correction to the potential,

$$\delta V = -\rho'(0)\,4\pi\,Z\alpha\,\delta^{(3)}(r), \tag{5}$$

and to the energy level of a hydrogenic system,

$$E^{(4)}_{\rm fns} = \langle\phi|\delta V|\phi\rangle = \frac{2\pi}{3}\,Z\alpha\,\langle r^2\rangle\,\phi^2(0), \tag{6}$$

where, for nS states,

$$\phi^2_{nS}(0) = \langle\delta^{(3)}(r)\rangle = \frac{(\mu Z\alpha)^3}{\pi n^3}, \tag{7}$$

and $\mu = m/(1+m/M)$ is the reduced mass of the atom.

To establish the importance of higher-order effects, we will need numerical values of the leading finite nuclear size effect in hydrogen and deuterium. The corresponding results, obtained assuming $r_p = 0.840\,87$ fm and $r_d = 2.125\,62$ fm, are, for the electronic atoms,

$$E^{(4)}_{\rm fns}(2S-1S,H) = -1\,368\,396\,r_p^2 = \;\;-9\,67\,541\ \text{Hz}, \tag{8}$$

$$E^{(4)}_{\rm fns}(2S-1S,D) = -1\,369\,513\,r_d^2 = -6\,187\,818\ \text{Hz}, \tag{9}$$

and for the muonic atoms,

$$E^{(4)}_{\rm fns}(2P-2S,\mu H) = -5.197\,45\,r_p^2 = \;\;-3.674\,92\ \text{meV}, \tag{10}$$

$$E^{(4)}_{\rm fns}(2P-2S,\mu D) = -6.073\,18\,r_d^2 = -27.440\,22\ \text{meV}. \tag{11}$$

We also observe that one of the relativistic $O(\alpha^2)$ corrections comes from the third term in Eq. (2),

$$\delta^{(2)}V = \tfrac{1}{2}\,\rho''(0)\,4\pi\,Z\alpha\,\nabla^2\delta^{(3)}(r). \tag{12}$$

Since its expectation value on nS states is singular, we will use dimensional regularization and combine this part with other $O(\alpha^2)$ corrections to obtain a finite result.

III. TWO-PHOTON EXCHANGE NUCLEAR STRUCTURE: MUONIC ATOMS

In this section we address the leading $O(\alpha)$ nuclear structure contribution $E^{(5)}$ in muonic atoms, which originates from the two-photon exchange between the bound lepton and the nucleus.

The elastic part $E^{(5)}_{\rm fns}$ can be obtained from the forward two-photon scattering amplitude at zero momentum,

$$E^{(5)}_{\rm fns} = \phi^2(0)\int\frac{d^3q}{(2\pi)^3}\,\mathrm{Tr}\Big[\gamma^0\,V\,\frac{1}{\not p - m}\,\gamma^0\,V\,\frac{(I+\gamma^0)}{4}\Big], \tag{13}$$

with $V = -4\pi\,Z\alpha/q^2$ and $p = (m,\vec q)$. This leads to the so-called Friar correction [9],

$$E^{(5)}_{\rm fns} = -(4\pi\,Z\alpha)^2\,\phi^2(0)\,2\,m\int\frac{d^3q}{(2\pi)^3}\,\frac{\rho^2(q^2)-1-2\,q^2\,\rho'(0)}{q^6}$$
$$= -\frac{\pi}{3}\,\phi^2(0)\,(Z\alpha)^2\,m\,r_Z^3, \tag{14}$$

where

$$r_Z^3 = \int d^3r_1\int d^3r_2\,\rho(r_1)\,\rho(r_2)\,|\vec r_1 - \vec r_2|^3. \tag{15}$$

As pointed out in Refs. [4,5], it is important to consider the Friar correction $E^{(5)}_{\rm fns}$ together with the corresponding inelastic part, because of a cancellation between them, occurring both for the muonic and the ordinary atoms. For this reason, we do not separate out $E^{(5)}_{\rm fns}$ but absorb it in the total nuclear structure correction $E^{(5)}$.

A. Muonic hydrogen

The inelastic two-photon exchange correction in μH has been extensively studied in the literature (see Ref. [10] and references therein). It is also given by the forward scattering amplitude and can be parameterized in terms of two spin-independent structure functions of the proton. Using dispersion relations, these functions are usually expressed in terms of the cross section of the inelastic photon scattering off the proton, which is extracted from experiment. The main problem of this approach is that one of the dispersion relations involves subtractions that can only be obtained from theory, and this introduces the dominant uncertainty.

There is good agreement between different calculations of the two-photon exchange correction, with the final result of $E^{(5)}(2P_{1/2}\!-2S,\mu\mathrm{H}) = E^{(5)}_{\rm fns} + E^{(5)}_{\rm pol} = 0.033\,2(20)$ meV assumed by the CREMA collaboration [11] in their determination of the proton charge radius. It is convenient to parametrize this result in terms of an effective radius r_{pF}, in analogy to Eq. (14),

$$E^{(5)}(\mu\mathrm{H}) = -\frac{\pi}{3}\,\phi^2(0)\,(Z\,\alpha)^2\,m\,r_{pF}^3, \tag{16}$$

with

$$r_{pF}^3 = 3.270\,(197)\ \mathrm{fm}^3. \tag{17}$$

This parametrization will be used below in our calculation of the inelastic contribution in other muonic atoms; see Eq. (24).

B. Muonic atoms other than hydrogen

For all nuclei other than the proton, the inelastic contribution is dominated by the electric dipole polarizability. For muonic atoms, one may assume the nonrelativistic approximation, so the second-order correction due to the electric dipole nuclear excitation is

$$E^{(5)}_{\mathrm{pol0}} = \alpha^2\,\langle\phi\,\phi_N|\frac{\vec d\cdot\vec r}{r^3}\,\frac{1}{E_N+E_0-H_N-H_0}\,\frac{\vec d\cdot\vec r}{r^3}\,|\phi\,\phi_N\rangle, \tag{18}$$

where $\vec d$ is the electric dipole operator divided by the elementary charge, and H_0 and H_N are the nonrelativistic Coulomb Hamiltonian for the muon and the nucleus, respectively. To the leading order in α, one may neglect the Coulomb interaction and replace $\phi(r)\to\phi(0)$ to obtain a compact formula for the leading two-photon exchange contribution,

$$E^{(5)}_{\mathrm{pol0}} = -\frac{4\,\pi\,\alpha^2}{3}\,\phi^2(0)\langle\phi_N|\vec d\,\sqrt{\frac{2\,m}{H_N-E_N}}\,\vec d\,|\phi_N\rangle, \tag{19}$$

which contributes 1.910 meV to the $2P-2S$ transition energy in muonic deuterium [6].

There are many corrections to the leading contribution [5–8], the most interesting of them being the one that partially cancels the Friar correction. To show this, following Ref. [5], we consider the muonic matrix element P for the nonrelativistic two-photon exchange,

$$P = \sum_{i,j}\langle\phi|\frac{\alpha}{|\vec r-\vec R_i|}\,\frac{1}{(H_0-E_0+E)}\,\frac{\alpha}{|\vec r-\vec R'_j|}|\phi\rangle, \tag{20}$$

where H_0 is the nonrelativistic Hamiltonian for the muon (electron) in the nonrecoil limit, and $\vec R_i$ is a position of the ith proton with respect to the nuclear mass center. Using the on-mass-shell approximation, subtracting the leading Coulomb interaction, the finite nuclear size, and the electric dipole polarizability, and expanding in the small parameter $\sqrt{2\,m\,E}|\vec R_i-\vec R'_j|$, we obtain

$$\begin{aligned} P &= \alpha^2\,\phi^2(0)\int\frac{d^3q}{(2\,\pi)^3}\left(\frac{4\,\pi}{q^2}\right)^2\left(E+\frac{q^2}{2\,m}\right)^{-1}\\ &\quad\times\left[e^{i\,\vec q\cdot(\vec R-\vec R')}-1+\frac{q^2}{6}\,(\vec R-\vec R')^2\right]\\ &\approx\sum_{i,j}\frac{\pi}{3}\,m\,\alpha^2\,\phi^2(0)\,|\vec R_i-\vec R'_j|^3\\ &\quad\times\left(1-\tfrac{1}{5}\,\sqrt{2\,m\,E}|\vec R_i-\vec R'_j|+\dots\right). \end{aligned} \tag{21}$$

The corresponding correction to the atomic energy is

$$E^{(5)}_{\mathrm{pol1}} = -\sum\!\!\!\!\!\!\int dE\int d^3R\,d^3R'\,\phi^*_N(\vec R)\,\phi_E(\vec R)\,\phi^*_E(\vec R')\,\phi_N(\vec R')\,P. \tag{22}$$

Let us consider only the first, E-independent term. When $\phi_E=\phi_N$, it corresponds to the elastic part, namely, the Friar correction given by Eq. (14). However, the inclusion of all excited states leads to

$$E^{(5)}_{\mathrm{pol1}} = -\frac{\pi}{3}\,m\,\alpha^2\,\phi^2(0)\sum_{i,j=1}^{Z}\langle\phi_N||\vec R_i-\vec R_j|^3|\phi_N\rangle, \tag{23}$$

which is much different from Eq. (14), in particular it vanishes for deuterium.

There are further nuclear polarizability corrections which were extensively studied in the literature [5–8]. It is convenient to write the final result for the two-photon exchange nuclear structure correction separating out the contribution due to the two-photon exchange with individual nucleons,

$$E^{(5)} = E^{(5)}_{\mathrm{pol}} - \frac{\pi}{3}\,m\,\alpha^2\,\phi^2(0)\left[Z\,r_{pF}^3+(A-Z)\,r_{nF}^3\right], \tag{24}$$

where $E^{(5)}_{\mathrm{pol}} = E^{(5)}_{\mathrm{pol0}} + E^{(5)}_{\mathrm{pol1}} + \dots$. Such representation of the nuclear structure correction is particularly advantageous for calculating isotope shifts, since the individual nucleon contributions partially cancel each other in the difference, together with the corresponding uncertainties. Calculating the two-photon exchange with individual nucleons, we take the effective proton radius r_{pF}^3 from Eq. (17), whereas for the neutron we assume the corresponding parameter to be four times smaller than that of the proton, $r_{nF}^3 = r_{pF}^3/4$, with uncertainty of 100%. This choice of r_{nF}^3 is in agreement with results summarized in Ref. [12] but anyhow requires further investigations.

Despite the fact that the literature results for $E^{(5)}_{\mathrm{pol}}$ in μD reported by different groups [5–8] are in good agreement with each other (see a summary in Ref. [12]), one should bear in mind that a number of higher-order effects exist that have not yet been addressed in any of the previous studies. Specifically, it has not so far been possible to include nucleon relativistic corrections to the coupling of the nucleus to the electromagnetic field. We thus believe that all theoretical predictions of $E^{(5)}_{\mathrm{pol}}$ in μD should bear an uncertainty whose relative value is approximately the ratio of the average nucleon binding energy to the nucleon mass, which is about 1%.

Summarizing our analysis of the existing literature results, we adopt the sum of entries $p_1\dots p_{12}$ labeled as "Our choice" in Table 3 in Ref. [12] as currently the best value of the two-photon nuclear polarizability correction to the $2P_{1/2}$–$2S$ transition energy in μD, and ascribe the uncertainty of 1% to it,

$$E^{(5)}_{\mathrm{pol}}(2P_{1/2}\text{–}2S,\mu D) = 1.6625\,(166)\ \mathrm{meV}. \tag{25}$$

The above uncertainty of $E^{(5)}_{\mathrm{pol}}$ is about 50% larger than the corresponding estimate of ±0.0107 meV given in Table 3 of Ref. [12]. Finally, we add the individual nucleon part in Eq. (24) and obtain the total two-photon nuclear structure correction to the $2P_{1/2}$–$2S$ transition energy in μD,

$$E^{(5)}(2P_{1/2}\text{–}2S,\mu D) = 1.7110\,(194)\ \mathrm{meV}, \tag{26}$$

which almost coincides with the corresponding result of 1.7096 (200) meV from Ref. [12], as given by Eq. (17) of that work.

IV. TWO-PHOTON EXCHANGE NUCLEAR STRUCTURE: ELECTRONIC ATOMS

The elastic (finite nuclear size) part of the two-photon exchange nuclear structure correction for electronic atoms is given by the same formula as for the muonic atoms, Eq. (13).

A. Electronic hydrogen

We calculate the elastic part of the nuclear structure correction for hydrogen according to Eq. (13) and using the result for the third Zemach moment from Ref. [13] obtained by averaging values measured in scattering experiments,

$$r_{pZ} = 1.587\,(26)\,r_p. \tag{27}$$

The corresponding result for the $2S$-$1S$ transition is

$$E^{(5)}_{\rm fns}(2S\text{–}1S,e\text{H}) = 0.0307\,(15)\ \text{kHz}. \tag{28}$$

The inelastic part of the two-photon exchange nuclear structure correction $E^{(5)}_{\rm pol}$ was derived in the logarithmic approximation in Ref. [14],

$$E^{(5)}_{\rm pol}(2S\text{–}1S,e\text{H}) = -m\,\alpha\,\phi^2(0)\,[5\,\alpha_p - \beta_p]\,\ln\frac{\bar{E}_p}{m}, \tag{29}$$

where $\bar{E}_p$ is the average proton excitation energy and α_p and β_p are the static proton polarizabilities extracted from experiment. Using the same average proton excitation energy $\bar{E}_p = 410$ MeV as in Ref. [14] and the updated results for the proton polarizabilities [15],

$$\begin{aligned}\alpha_p &= 10.65\,(35)(20)(30)\ \text{fm}^3,\\ \beta_p &= 3.15\,(35)(20)(30)\ \text{fm}^3,\end{aligned} \tag{30}$$

we obtain the result for the $2S$-$1S$ transition of

$$E^{(5)}_{\rm pol}(2S\text{–}1S,e\text{H}) = 0.0567\,(85)\ \text{kHz}, \tag{31}$$

where, following Ref. [14], we assumed a 15% uncertainty due to the leading logarithmic approximation.

The total result for the two-photon nuclear structure correction in electronic hydrogen is

$$E^{(5)}(2S\text{–}1S,e\text{H}) = E^{(5)}_{\rm fns} + E^{(5)}_{\rm pol} = 0.0874\,(86)\ \text{kHz}, \tag{32}$$

which could be compared with the corresponding result of 0.091 (11) kHz from Ref. [16].

B. Electronic atoms other than hydrogen

Similar to the muonic atoms, it is convenient to write the total two-photon exchange nuclear structure correction separating out the contribution due to the interaction with individual nucleons,

$$\begin{aligned}E^{(5)} = E^{(5)}_{\rm pol} - m\,\alpha\,\phi^2(0)\bigg\{&\frac{\pi}{3}\,\alpha\,Z\,r^3_{pZ} + Z\,[5\,\alpha_p - \beta_p]\,\ln\frac{\bar{E}_p}{m}\\ &+ (A-Z)\,[5\,\alpha_n - \beta_n]\,\ln\frac{\bar{E}_n}{m}\bigg\}.\end{aligned} \tag{33}$$

In the above formula, the first and the second terms in the brackets represent the elastic and the inelastic interactions with individual protons, respectively, whereas the third term comes from the inelastic interaction with individual neutrons. The parameters for the protons are the same as for hydrogen, whereas for the neutrons we use the experimental polarizabilities [15],

$$\begin{aligned}\alpha_n &= 11.55\,(125)(20)(80)\ \text{fm}^3,\\ \beta_n &= 3.65\,(125)(20)(80)\ \text{fm}^3,\end{aligned} \tag{34}$$

and the same value of $\bar{E}_n = 410$ MeV as for the proton. We note that the elastic interaction of the bound electron with the nucleus as a whole is absorbed in $E^{(5)}_{\rm pol}$, reflecting the fact that the third Zemach moment correction for a compound nucleus largely cancels out between the elastic and inelastic parts in the same way as in muonic atoms.

Similar to the muonic atoms, the nuclear polarizability correction $E^{(5)}_{\rm pol}$ in Eq. (33) comes from the electric dipole polarizability, which, however, takes a very different form for the electronic atoms. Since in this case the nonrelativistic approximation is not valid, one should consider the complete two-photon exchange and keep the relativistic form of the matrix elements,

$$\begin{aligned}E^{(5)}_{\rm pol} &= i\,e^2\,\phi^2(0)\int\frac{d\,\omega}{2\,\pi}\int\frac{d^3k}{(2\,\pi)^3}\,\omega^2\\ &\quad\times\frac{\left(\delta^{ik} - \frac{k^i\,k^k}{\omega^2}\right)}{\omega^2 - k^2}\,\frac{\left(\delta^{jl} - \frac{k^j\,k^l}{\omega^2}\right)}{\omega^2 - k^2}\\ &\quad\times\text{Tr}\left[\left(\gamma^j\,\frac{1}{\not p - \not k - m}\gamma^i + \gamma^i\,\frac{1}{\not p + \not k - m}\,\gamma^j\right)\frac{(\gamma^0 + I)}{4}\right]\\ &\quad\times\langle\phi_N|d^k\,\frac{1}{E_N - H_N - \omega}\,d^l|\phi_N\rangle + \ldots\\ &= E^{(5)}_{\rm pol1} + E^{(5)}_{\rm pol2} + E^{(5)}_{\rm pol3} + \ldots,\end{aligned} \tag{35}$$

where $p = (m,\vec{0})$. Assuming that the electron mass is much smaller than the nuclear excitation energy, the leading nuclear polarizability correction becomes

$$E^{(5)}_{\rm pol1} = -m\,\alpha^2\,\phi^2(0)\,\frac{2}{3}\,\langle\phi_N|\vec{d}\,\frac{1}{H_N - E_N}\left[\frac{19}{6} + 5\,\ln\frac{2\,(H_N - E_N)}{m}\right]\vec{d}\,|\phi_N\rangle. \tag{36}$$

The corresponding contribution to the $2S$-$1S$ transition in ordinary deuterium is 19.26 (6) kHz [17].

Various small corrections to the electric dipole polarizability for electronic atoms were considered by Friar in Ref. [4]. In particular, it was shown there that the Zemach contribution for deuterium vanishes in the same way as for the muonic deuterium. Furthermore, the higher-order terms in the $m/(H_N - E_N)$ expansion of Eq. (35) give rise to a correction,

$$E^{(5)}_{\rm pol2} = -m^3\,\alpha^2\,\phi^2(0)\,\frac{2}{3}\,\langle\phi_N|\vec{d}\,\frac{1}{(H_N - E_N)^3}\left[-\frac{283}{80} + \frac{15}{4}\,\ln\frac{2\,(H_N - E_N)}{m}\right]\vec{d}|\phi_N\rangle, \tag{37}$$

which contributes 0.106 kHz to the $2S$-$1S$ transition in ordinary deuterium [4]. Another important correction is the one due to the magnetic suceptibility [4],

$$E^{(5)}_{\rm pol3} = m\,\alpha^2\,\phi^2(0)\,\frac{2}{3}\,\langle\phi_N|\vec\mu\,\frac{1}{(H_N-E_N)'}\left[-\frac{1}{6}+\ln\frac{2\,(H_N-E_N)}{m}\right]\vec\mu\,|\phi_N\rangle, \tag{38}$$

where μ is the magnetic moment operator divided by an elementary charge. It leads to a correction of $-0.307(2)(6)$ kHz to the $2S$-$1S$ transition in eD [4].

There were further corrections to the electric dipole polarizability considered in Ref. [4]. However, we are convinced that they were not treated correctly and, moreover, that there are many more relativistic corrections of the same order. For this reason we disregard the additional corrections from Ref. [4] and assume the total polarizability correction to be the sum of Eqs. (36), (37), and (38). Specifically, the result for the nuclear polarizability to the $2S$-$1S$ transition in electronic deuterium is

$$E^{(5)}_{\rm pol}(2S\text{–}1S,D) = 19.06\,(20)\,\text{kHz}. \tag{39}$$

Adding the individual nucleon part contribution of 0.149 (22) kHz, we obtain the total two-photon exchange nuclear structure contribution of

$$E^{(5)}(2S\text{–}1S,D) = 19.21\,(20)\,\text{kHz}, \tag{40}$$

which could be compared with the sum of the nuclear polarizability correction and the third Zemach contribution from Ref. [16], $18.70\,(7)+0.51 = 19.21\,(7)$ kHz, perfect agreement of the numerical values being probably accidental.

V. THREE-PHOTON EXCHANGE ELASTIC CONTRIBUTION

This contribution has been studied by different methods and a number of authors, of note analytically by Friar in Ref. [9], and numerically by solving the Dirac equation in the field of finite size nucleus [18]. Here we present an alternative analytical approach, which leads to much simpler analytic formulas. A numerical verification of our formulas is given in Appendix B.

In the standard analytic approach, one applies the perturbation theory to the Dirac energies with the perturbing potential $\delta V = V - V_0$, where $V_0 = -Z\alpha/r$ and V is the Coulomb potential from the finite-size nucleus,

$$\delta^{(1)}E = \langle\bar\psi|\delta V|\psi\rangle, \tag{41}$$

$$\delta^{(2)}E = \langle\bar\psi|\delta V\,\frac{1}{(\not p-\gamma^0\,V_0-m)'}\,\delta V|\psi\rangle, \tag{42}$$

$$\delta^{(3)}E = \langle\bar\psi|\delta V\,\frac{1}{(\not p-\gamma^0\,V_0-m)'}\,(\delta V-\langle\delta V\rangle) \times\frac{1}{(\not p-\gamma^0\,V_0-m)'}\,\delta V|\psi\rangle. \tag{43}$$

One can use the exact Dirac wave function and the reduced Dirac propagators to calculate the $O(\alpha^2)$ correction to the finite nuclear size [9], which we call the elastic three-photon exchange correction. However, we will not use the above formulas but employ a different approach, which we call the scattering amplitude approach. In this approach, the $O(\alpha^2)$ relativistic correction to the finite nuclear size is induced by the elastic three photon exchange. The corresponding correction can be divided into the low and the high energy momentum exchange parts, $E^{(6)}_{\rm fns} = E_L + E_H$. These parts are calculated as follows.

A. Three-photon exchange: Low-energy part

The low-energy part E_L is again split into two parts

$$E_L = E_{L1} + E_{L2}, \tag{44}$$

$$E_{L1} = \langle\delta V\,\frac{1}{(E-H)'}\,\delta V\rangle + \langle\delta^{(2)}V\rangle, \tag{45}$$

$$E_{L2} = \langle\phi|\frac{1}{8\,m^2}\,\nabla^2(\delta V) + \frac{1}{4\,m^2}\,\vec\sigma\cdot\vec\nabla(\delta V)\times\vec p|\phi\rangle + 2\,\langle\phi|\delta V\frac{1}{(E-H)'}\left[-\frac{p^4}{8\,m^3}+\frac{\pi\,Z\alpha}{2\,m^2}\,\delta^3(r)\right]|\phi\rangle, \tag{46}$$

where E_{L1} is the nonrelativistic contribution proportional to r_C^4, and E_{L2} is the relativistic part proportional to r_C^2. All these matrix elements are calculated in $d = 3-2\,\epsilon$ dimensions. The following results are obtained for the nS states:

$$E_{L1}(nS) = [\rho'(0)]^2\,\frac{16\,(Z\alpha)^6}{n^3}\left[-\frac{1}{n}-\frac{1}{2}+\gamma-\ln\frac{n}{2}+\Psi(n)\right] + (Z\alpha)^3\left[-\frac{1}{\epsilon}+4\,\ln(Z\alpha)\right]4\,[\rho'(0)]^2\,\langle\pi\,\delta^{(d)}(r)\rangle + \frac{4\,(Z\alpha)^6\,\rho''(0)}{n^5}, \tag{47}$$

$$E_{L2}(nS) = \rho'(0)\,\frac{4\,(Z\alpha)^6}{n^3}\left[\frac{9}{4n^2}-\frac{1}{n}-\frac{5}{2}+\gamma-\ln\frac{n}{2}+\Psi(n)\right] + (Z\alpha)^3\left[-\frac{1}{\epsilon}+4\,\ln(Z\alpha)\right]\rho'(0)\,\langle\pi\,\delta^{(d)}(r)\rangle, \tag{48}$$

and for the nP states,

$$E_L(nP_{1/2}) = (Z\alpha)^6\left[-\frac{9}{4}\,\rho'(0)+3\,\rho''(0)\right]R'_{n1}(0)^2, \tag{49}$$

$$E_L(nP_{3/2}) = (Z\alpha)^6\,3\,\rho''(0)\,R'_{n1}(0)^2, \tag{50}$$

where

$$R'_{n1}(0)^2 = \frac{4}{9\,n^3}\left(1-\frac{1}{n^2}\right). \tag{51}$$

For all higher-L states E_L vanishes.

B. Three-photon exchange: High-energy part

We start by introducing the two potentials in d-dimensions that will appear in the evaluation of the high energy-part E_H,

$$V_d(r) = 4\pi \int \frac{d^d q}{(2\pi)^d}\, e^{i\vec{q}\cdot\vec{r}}\, \frac{\rho(q^2)}{q^2}, \tag{52}$$

$$V_d^{(2)}(r) = 4\pi \int \frac{d^d q}{(2\pi)^d}\, e^{i\vec{q}\cdot\vec{r}}\, \frac{\rho(q^2)}{q^4}. \tag{53}$$

Their large r asymptotics are

$$V_d(r) = \mathcal{V}(r) + \text{local terms}, \tag{54}$$

$$V_d^{(2)}(r) = \mathcal{V}^{(2)}(r) + \rho'(0)\,\mathcal{V}(r) + \text{local terms}, \tag{55}$$

where $\mathcal{V}(r)$ and $\mathcal{V}^{(2)}(r)$ are defined by Eqs. (A2)–(A5) and in $d = 3$,

$$V(r) = \frac{1}{r} + \text{local terms}, \tag{56}$$

$$V^{(2)}(r) = -\frac{r}{2} + \frac{\rho'(0)}{r} + \text{local terms}, \tag{57}$$

where the local terms vanish outside the nucleus.

Now we proceed to the derivation of the high-energy part E_H. It is given by the three-photon scattering amplitude with momenta $p_i = (m,\vec{q}_i)$,

$$E_H = -(4\pi\, Z\alpha)^3\, \phi^2(0) \int \frac{d^d q_1}{(2\pi)^d} \int \frac{d^d q_2}{(2\pi)^d}\, \frac{\rho(q_1^2)}{q_1^4}\, \frac{\rho(q_2^2)}{q_2^4}\, \frac{\rho[(\vec{q}_1-\vec{q}_2)^2]}{(\vec{q}_1-\vec{q}_2)^2} \mathrm{Tr}\left[(\not p_1 + m)\,\gamma_0\,(\not p_2 + m)\,\frac{(\gamma_0 + I)}{4}\right]. \tag{58}$$

The above trace equates to $4\,m^2 + \vec{q}_1\,\vec{q}_2$, so we can split E_H into the nonrelativistic and relativistic parts,

$$E_H = E_{H1} + E_{H2}. \tag{59}$$

The nonrelativistic part E_{H1} is

$$\begin{aligned} E_{H1} &= -(4\pi\, Z\alpha)^3\, \phi^2(0)\, 4\,m^2 \int \frac{d^d q_1}{(2\pi)^d} \int \frac{d^d q_2}{(2\pi)^d}\, \frac{\rho(q_1^2)}{q_1^4}\, \frac{\rho(q_2^2)}{q_2^4}\, \frac{\rho[(\vec{q}_1-\vec{q}_2)^2]}{(\vec{q}_1-\vec{q}_2)^2} \\ &= -\phi^2(0)\,(Z\alpha)^3\, 4\,m^2 \int d^d r\, V_d(r)\,\big[V_d^{(2)}(r)\big]^2. \end{aligned} \tag{60}$$

To calculate this integral, we split the integration region into $r < \Lambda$ and $r \geqslant \Lambda$. The first integral is finite in $d = 3$ but diverges at large Λ, and in the second integral one can use the asymptotic form of potentials,

$$\begin{aligned} E_{H1} &= -\phi^2(0)\,(Z\alpha)^3\, 4\,m^2 \left\{4\pi \int^{\Lambda} dr\, r^2\, V(r)\,[V^{(2)}(r)]^2 + \int_{\Lambda} d^d r\, \mathcal{V}(r)\,[\mathcal{V}^{(2)}(r) + \rho'(0)\,\mathcal{V}(r)]^2\right\} \\ &= \phi^2(0)\,(Z\alpha)^3\, 4\,m^2\, 4\pi \int^{\infty} dr\, \ln(r/r_C)\, \frac{d}{dr}\, r^3 \left\{V(r)\,[V^{(2)}(r)]^2 - \frac{1}{r}\left[\frac{r}{2} - \frac{\rho'(0)}{r}\right]^2\right\} \\ &\quad -\phi^2(0)\,(Z\alpha)^3\, 4\,m^2\, \rho'(0)^2 \left\{4\pi\, \ln(\Lambda/r_C) + \int_{\Lambda} d^d r\, \mathcal{V}(r)^3\right\}. \end{aligned} \tag{61}$$

The expression under the first integral is a local function of r, so this integral is effectively over the nuclear size, which allows us to introduce an effective nuclear radius r_{C1} as

$$\ln\frac{r_{C1}}{r_C} + 2 = \frac{36}{r_C^4} \int^{\infty} dr\, \ln(r/r_C)\, \frac{d}{dr}\, r^3 \left\{V(r)\,[V^{(2)}(r)]^2 - \frac{1}{r}\left[\frac{r}{2} - \frac{\rho'(0)}{r}\right]^2\right\}. \tag{62}$$

So, the first $O(\alpha^2)$ correction E_{H1} is represented in the following form:

$$E_{H1} = 4\pi\, \phi^2(0)\,(Z\alpha)^3\, 4\,m^2\, \frac{r_C^4}{36}\left[\frac{1}{4\epsilon} + \frac{5}{2} + \gamma + \ln(r_{C1}\, m)\right]. \tag{63}$$

The relativistic part E_{H2} is

$$E_{H2} = -(4\pi\, Z\alpha)^3\, \phi^2(0) \int \frac{d^d q_1}{(2\pi)^d} \int \frac{d^d q_2}{(2\pi)^d}\, \frac{\rho(q_1^2)}{q_1^4}\, \frac{\rho(q_2^2)}{q_2^4}\, \frac{\rho[(\vec{q}_1-\vec{q}_2)^2]}{(\vec{q}_1-\vec{q}_2)^2}\, \vec{q}_1\vec{q}_2$$
$$= \phi^2(0)\,(Z\alpha)^3 \int d^d r\, \{2\pi\,\rho(r)\, V_d^{(2)}(r) - [V_d(r)]^2\}\, V_d^{(2)}(r), \tag{64}$$

and we proceed in a similar way as in the case of E_{H1}, namely,

$$E_{H2} = \phi^2(0)\,(Z\alpha)^3 \left\{4\pi \int^{\Lambda} dr\, r^2 \Big[2\pi\,\rho(r)\, V^{(2)}(r) - [V(r)]^2\Big]\, V^{(2)}(r) - \int_{\Lambda} d^d r [\mathcal{V}(r)]^2\, [\mathcal{V}^{(2)}(r) + \rho'(0)\,\mathcal{V}(r)]\right\}$$
$$= -\phi^2(0)\,(Z\alpha)^3\, 4\pi \int^{\infty} dr\, \ln(r/r_C) \frac{d}{dr}\, r^3 \left\{2\pi\,\rho(r)\,[V^{(2)}(r)]^2 - [V(r)]^2\, V^{(2)}(r) - \frac{1}{r^2}\left[\frac{r}{2} - \frac{\rho'(0)}{r}\right]\right\}$$
$$-\,\phi^2(0)\,(Z\alpha)^3\,\rho'(0) \left\{4\pi\, \ln(\Lambda/r_C) + \int_{\Lambda} d^d r\, [\mathcal{V}(r)]^3\right\}. \tag{65}$$

The expression under the first integral is a local function, so we can introduce the second effective nuclear radius r_{C2} as

$$\ln \frac{r_{C2}}{r_C} - 1 = \frac{6}{r_C^2} \int^{\infty} dr\, \ln(r/r_C) \frac{d}{dr}\, r^3 \left\{2\pi\,\rho(r)\,[V^{(2)}(r)]^2 - [V(r)]^2\, V^{(2)}(r) - \frac{1}{r^2}\left[\frac{r}{2} - \frac{\rho'(0)}{r}\right]\right\}. \tag{66}$$

So, the second $O(\alpha^2)$ correction is given by

$$E_{H2} = -4\pi\,(Z\alpha)^3\,\phi^2(0)\,\frac{r_C^2}{6}\left[\frac{1}{4\epsilon} - \frac{1}{2} + \gamma + \ln(r_{C2}\, m)\right]. \tag{67}$$

C. Three-photon elastic exchange: Total result

The complete $O(\alpha^2)$ finite nuclear size correction for an arbitrary nucleus is given by the sum $E_{\rm fns}^{(6)} = E_L + E_H = E_{L1} + E_{L2} + E_{H1} + E_{H2}$, with the result

$$E_{\rm fns}^{(6)}(nS) = -(Z\alpha)^6\, m^3\, r_C^2\, \frac{2}{3\,n^3}\left[\frac{9}{4n^2} - 3 - \frac{1}{n} + 2\gamma - \ln\frac{n}{2} + \Psi(n) + \ln(m\, r_{C2}\, Z\alpha)\right]$$
$$+\,(Z\alpha)^6\, m^5\, r_C^4\, \frac{4}{9\,n^3}\left[-\frac{1}{n} + 2 + 2\gamma - \ln\frac{n}{2} + \Psi(n) + \ln(m\, r_{C1}\, Z\alpha)\right] + (Z\alpha)^6\, m^5\, r_{CC}^4\, \frac{1}{15\,n^5}, \tag{68}$$

$$E_{\rm fns}^{(6)}(nP_{1/2}) = (Z\alpha)^6\, m \left(\frac{m^2\, r_C^2}{6} + \frac{m^4\, r_{CC}^4}{45}\right) \frac{1}{n^3}\left(1 - \frac{1}{n^2}\right), \tag{69}$$

$$E_{\rm fns}^{(6)}(nP_{3/2}) = (Z\alpha)^6\, m^5\, r_{CC}^4\, \frac{1}{45\, n^3}\left(1 - \frac{1}{n^2}\right), \tag{70}$$

$$E_{\rm fns}^{(6)}(nL_J) = 0 \text{ for } L > 1, \tag{71}$$

where $r_{CC}^4 = \langle r^4 \rangle$ and the effective nuclear charge radii r_{C1} and r_{C2} defined by Eqs. (62) and (66) encode the high-momentum contributions and are expected to be of the order of r_C. Equations (68)–(71) are valid both for electronic and muonic atoms. However, in the case of the electronic atoms, the terms proportional to r_C^4 and r_{CC}^4 in these formulas are smaller than the next-order correction and thus should be neglected.

Equations (68)–(71) depend on the nuclear model through the effective nuclear charge radii r_{C1} and r_{C2}. However, as we demonstrate below, the terms with r_{C1} and r_{C2} exactly cancel in the sum with the corresponding inelastic contribution, so their model dependence is irrelevant for the final result.

Table I presents our results for the effective charge radii r_{C1} and r_{C2}, for two models of the nuclear charge distribution. The ratios listed in the table are proven to be independent of the nuclear model parameters λ and a, thus making the corresponding results valid for an arbitrary nucleus and both for the electronic and muonic atoms.

The formulas for $E_{\rm fns}^{(6)}$ have been derived in the nonrecoil limit, i.e., assuming the infinite nuclear mass. This is different from the approach by Friar in Ref. [9], in which he replaced the lepton mass by the reduced mass of the system. We do not think such a replacement is valid. However, recoil corrections for muonic atoms are significant and can be partially accounted for by the $(\mu/m)^3$ scaling factor that comes from the square of the nonrelativistic wave function at origin.

Equations (68)–(71) can be compared with the analytical results by Friar derived within a different approach [9]. However, the formulas of Ref. [9] are so complicated that a direct comparison is not possible, except for the state dependence which is in perfect agreement. A comparison of numerical results presented in Sec. VIII shows a reasonable but not perfect agreement. To verify our formulas, we performed a high-precision numerical calculation, by solving the Dirac

TABLE I. Various results for the exponential and the Gaussian models of the nuclear charge distributions, $r_C = \sqrt{\langle r^2\rangle}$, $r_{CC} = \sqrt[4]{\langle r^4\rangle}$, r_{C1} is defined in Eq. (62), r_{C2} in Eq. (66), r_Z in Eq. (15).

	Exponential	Gaussian
$\rho(q^2)$	$\frac{\lambda^4}{(\lambda^2+q^2)^2}$	$\exp(\frac{a\,q^2}{2})$
$\rho(r)$	$\frac{\lambda^3}{8\pi}\,e^{-\lambda\,r}$	$\frac{1}{(2\pi a)^{3/2}}\,\exp(-\frac{r^2}{2\,a})$
r_C	$\frac{2\sqrt{3}}{\lambda}$	$\sqrt{3\,a}$
$V(r)$	$\frac{1}{r}-\frac{e^{-\lambda r}}{r}-\frac{\lambda}{2}\,e^{-\lambda\,r}$	$\frac{1}{r}\,\mathrm{erf}(\frac{r}{\sqrt{2a}})$
$V^{(2)}(r)$	$-\frac{r}{2}-\frac{2}{\lambda^2 r}+\frac{1}{2\lambda}\,e^{-\lambda\,r}+\frac{2}{\lambda^2}\,\frac{e^{-\lambda r}}{r}$	$-\sqrt{\frac{a}{2\pi}}\,\exp(-\frac{r^2}{2a})-\frac{(a+r^2)}{2\,r}\,\mathrm{erf}(\frac{r}{\sqrt{2a}})$
r_{C1}/r_C	1.090 044	0.558 872
r_{C2}/r_C	1.068 497	1.014 281
r_{CC}/r_C	1.257 433	1.136 219
r_Z/r_C	1.558 965	1.514 599

equation numerically and identifying the $O(\alpha^2)$ finite nuclear size contribution, as described in Appendix B. Perfect agreement between analytical and numerical approaches confirms the correctness of Eqs. (68)–(71).

VI. INELASTIC THREE-PHOTON EXCHANGE CORRECTION IN MUONIC DEUTERIUM

The inelastic three-photon exchange nuclear structure correction has not yet been studied in the literature and is the main topic of this work. With momenta of order $q \sim \sqrt{2\,m\,\Lambda}$, which is much lower than the inverse of the nuclear size, the muon kinetic energy becomes comparable to the characteristic nuclear excitation energy Λ, and thus the muon starts to probe the nuclear structure and see individual nucleons. It means that the total correction does not involve contributions coming from muon momenta of the order of the inverse of nuclear size, and there is no place for the elastic high-energy parts encoded in r_{C1} and r_{C2} effective nuclear radii.

We represent the total nuclear structure correction $E^{(6)}$ as a sum of several parts,

$$E^{(6)} = E^{(6)}_1 + E^{(6)}_2 + E_C + E^{(6)}_{\rm np} = E^{(6)}_{\rm fns} + E^{(6)}_{1,\rm pol} + E^{(6)}_{2,\rm pol} + E_C + E^{(6)}_{\rm np}, \tag{72}$$

where the elastic part $E^{(6)}_{\rm fns} = E^{(6)}_{1,\rm fns} + E^{(6)}_{2,\rm fns}$ was calculated in the previous section, $E^{(6)}_1$ and $E^{(6)}_{1,\rm pol}$ are proportional to r_C^4, $E^{(6)}_2$ and $E^{(6)}_{2,\rm pol}$ are proportional to r_C^2, E_C is the known Coulomb distortion correction [5,7], and $E^{(6)}_{\rm np}$ is the contribution due to the inelastic interaction with individual nucleons.

Below we calculate $E^{(6)}_1$ and $E^{(6)}_2$. Because the two-photon exchange nuclear structure correction $E^{(5)}_{\rm pol}$ was previously shown to be dominated by the electric dipole types of the nuclear polarizability, we will assume that the same holds for the three-photon exchange nuclear structure correction.

A. Inelastic contribution $\propto R^2$

We represent the total $E^{(6)}_2$ correction as a sum of several parts,

$$E^{(6)}_2 = E_{L2} + E_C + E_R + E_{H2}(p), \tag{73}$$

calculated in the following. Let us consider the two radiative photon exchange between the muon and the nucleus, taking into account the Coulomb interaction V. The corresponding energy shift is

$$\delta E = i\,e^2 \int \frac{d\,\omega}{2\,\pi} \int \frac{d^3k_1}{(2\,\pi)^3} \int \frac{d^3k_2}{(2\,\pi)^3}\,\omega^2\,\frac{\left(\delta^{ik}-\frac{k_1^i\,k_1^k}{\omega^2}\right)}{\omega^2-k_1^2}\,\frac{\left(\delta^{jl}-\frac{k_2^j\,k_2^l}{\omega^2}\right)}{\omega^2-k_2^2}\Big[\langle\bar\psi|\gamma^j\,e^{i\,\vec k_2\vec r}\,\frac{1}{\not p-\gamma^0\,V-\gamma^o\,\omega-m}\gamma^i\,e^{i\,\vec k_1\vec r}|\psi\rangle + \langle\bar\psi|\gamma^i\,e^{i\,\vec k_1\vec r}\,\frac{1}{\not p-\gamma^o\,V+\gamma^0\,\omega-m}\,\gamma^j\,e^{i\,\vec k_2\vec r}\,|\psi\rangle\Big]\langle\phi_N|d^k\,\frac{1}{E_N-H_N-\omega}\,d^l|\phi_N\rangle, \tag{74}$$

where $\vec d$ is the dipole moment operator divided by the elementary charge. In the case of deuteron $\vec d$ is equal to the position of the proton with respect to the mass center $\vec d = \vec R$. In the nonrelativistic limit, δE takes the well known form of Eq. (16). The corresponding low-energy α^6 contribution is obtained from Eq. (74) by assuming that the muon momenta are of the order of $m\,\alpha$. Then $E_0 - H_0$ can be neglected in comparison to $E_N - H_N$ and one obtains (with $d = 3 - 2\,\epsilon$),

$$\delta_L E = \alpha^2\,\langle\phi|\frac{1}{r^4}|\phi\rangle_\epsilon\,\frac{1}{d}\,\langle\phi_N|\vec R\,\frac{1}{E_N-H_N}\,\vec R\,|\phi_N\rangle, \tag{75}$$

and

$$\langle\phi|\frac{1}{r^4}|\phi\rangle_\epsilon = \langle[\nabla\mathcal{V}(r)]^2\rangle = \left\langle\frac{1}{r^4}\right\rangle + \phi^2(0)\,4\,\pi\left(-\frac{1}{2\,\epsilon} + 2\,\ln\alpha + 2\right), \tag{76}$$

were $\langle 1/r^4\rangle$ is defined as an integral from a small radius a to infinity with the $1/a$ and $\ln a + \gamma$ terms subtracted out. The high-energy α^6 part is obtained by assuming that muon momenta are of the order $\sqrt{2\,m\,\Lambda}$. Then we can use the explicit Coulomb correction

$$\begin{aligned}\delta_H E = {}& -i\,e^2\int\frac{d\,\omega}{2\,\pi}\int\frac{d^3k_1}{(2\,\pi)^3}\int\frac{d^3k_2}{(2\,\pi)^3}\,\omega^2\,\frac{\left(\delta^{ik}-\frac{k_1^i\,k_1^k}{\omega^2}\right)}{\omega^2-k_1^2}\,\frac{\left(\delta^{jl}-\frac{k_2^j\,k_2^l}{\omega^2}\right)}{\omega^2-k_2^2}\,\frac{4\,\pi\,\alpha}{(\vec{k}_1+\vec{k}_2)^2}\\ &\times\mathrm{Tr}\bigg[\bigg(\gamma^j\,\frac{1}{\not p+\not k_2-m}\,\gamma^0\,\frac{1}{\not p-\not k_1-m}\,\gamma^i+\gamma^0\,\frac{1}{\not p-\not k_1-\not k_2-m}\,\gamma^j\,\frac{1}{\not p-\not k_1-m}\,\gamma^i\\ &+\gamma^j\,\frac{1}{\not p+\not k_2-m}\,\gamma^i\,\frac{1}{\not p+\not k_1+\not k_2-m}\gamma^0\bigg)\,\frac{(\gamma^0+I)}{4}\bigg]\,\phi^2(0)\\ &\times\left[\langle\phi_N|R^k\,\frac{1}{E_N-H_N-\omega}\,R^l|\phi_N\rangle+\langle\phi_N|R^l\,\frac{1}{E_N-H_N+\omega}\,R^k|\phi_N\rangle\right],\end{aligned} \tag{77}$$

where $k_1 = (\omega,\vec{k}_1)$, $k_2 = (-\omega,\vec{k}_1)$, and $p = (m,\vec{0})$. Assuming that $(H_N - E_N)/m$ is small, one performs an expansion and obtains

$$\begin{aligned}\delta_H E = {}& -\pi\,\alpha^3\,\phi^2(0)\,\frac{1}{d}\left\{\langle\phi_N|\vec{R}\frac{4}{H_N-E_N}\left[\frac{1}{2\,\epsilon}-1+\ln 2-\ln\frac{(H_N-E_N)}{m}\right]\vec{R}\,|\phi_N\rangle\right.\\ &\left.+\langle\phi_N|\vec{R}\left[\frac{1}{2\,\epsilon}+\frac{13}{2}-13\,\ln 2-5\,\ln\frac{(H_N-E_N)}{m}\right]\vec{R}\,|\phi_N\rangle\right\}.\end{aligned} \tag{78}$$

The sum of $\delta_L E$ in Eq. (75) and $\delta_H E$ in Eq. (78) gives the leading Coulomb distortion correction E_C,

$$E_C = -\frac{1}{3}\left\langle\frac{1}{r^4}\right\rangle\langle\phi_N|\vec{R}\,\frac{1}{H_N-E_N}\,\vec{R}\,|\phi_N\rangle - \frac{4\,\pi}{3}\,\phi^2(0)\,\langle\phi_N|\,\vec{R}\,\frac{1}{H_N-E_N}\left[1+\ln\!\left(\frac{2\,m\,\alpha^2}{H_N-E_N}\right)\right]\vec{R}\,|\phi_N\rangle, \tag{79}$$

where

$$\left\langle\frac{1}{r^4}\right\rangle_{nS} = \frac{8}{n^3}\left[-\frac{5}{3}+\frac{1}{2\,n}+\frac{1}{6\,n^2}+\gamma+\Psi(n)-\ln\frac{n}{2}\right], \tag{80}$$

$$\left\langle\frac{1}{r^4}\right\rangle_{nP} = \frac{2\,(3\,n^2-2)}{15\,n^5}, \tag{81}$$

and the relativistic correction E_R,

$$E_R = -\pi\,\alpha^3\,\phi^2(0)\,\frac{r_s^2}{6}\left(\frac{1}{\epsilon}+\frac{41}{3}-26\,\ln 2-10\,\ln\frac{\langle E\rangle_2}{m}\right), \tag{82}$$

where $r_s^2 = \langle R^2\rangle$ is the deuteron structure radius and

$$\ln\frac{\langle E\rangle_2}{m} = \frac{1}{r_s^2}\,\langle\phi|\vec{R}\,\ln\frac{(H_N-E_N)}{m}\,\vec{R}\,|\phi\rangle. \tag{83}$$

Although there is no elastic high-energy part, the individual proton contributes

$$E_{H2}(p) = -\pi\,\alpha^3\,\phi^2(0)\,\frac{r_p^2}{6}\left[\frac{1}{\epsilon}-2+4\,\gamma+4\,\ln(r_{p2}\,m)\right]. \tag{84}$$

The last contribution E_{L2} is exactly the same as the one of Eq. (48) with the deuteron radii.

Finally, the total nuclear structure contribution $E_2^{(6)} \propto R^2$ is given by the sum of the elastic and inelastic parts,

$$\begin{aligned}E_2^{(6)}(nS) = {}& E_C(nS) - \alpha^6\,m^3\,\frac{2}{3\,n^3}\left\{r_d^2\left[\frac{9}{4n^2}-3-\frac{1}{n}+2\,\gamma-\ln\frac{n}{2}+\Psi(n)+\ln\alpha\right]\right.\\ &\left.+r_s^2\left(\frac{47}{12}-\frac{13}{2}\,\ln 2-\frac{5}{2}\,\ln\frac{\langle E\rangle_2}{m}-\gamma\right)+r_p^2\,\ln(r_{p2}\,m)\right\}.\end{aligned} \tag{85}$$

It is remarkable that the part of the elastic contribution depending on the effective deuteron radius r_{d2} does not show up in total $E_2^{(6)}$. Separately, the expression for the inelastic contribution is

$$E_{2,\mathrm{pol}}^{(6)} = E_2^{(6)} - E_{2,\mathrm{fns}}^{(6)} - E_C$$
$$= -\frac{2\,\alpha^6\,m^3}{3\,n^3}\,\delta_{l0}\left\{r_s^2\left(\frac{47}{12} - \frac{13}{2}\,\ln 2 - \frac{5}{2}\,\ln\frac{\langle E\rangle_2}{m}\right) + r_p^2\,[\gamma + \ln(r_{p2}\,m)] - r_d^2\,[\gamma + \ln(r_{d2}\,m)]\right\}. \tag{86}$$

The averaged excitation energy $\langle E\rangle_2$ has been calculated using the well known AV18 potential [19] with the result

$$\langle E\rangle_2 = 7.37(7)\ \mathrm{MeV}, \tag{87}$$

where the 1% uncertainty is our guess for dependence on the potential model. $\langle E\rangle_2$ is exactly the same for μD and eD, because the lepton mass cancels out between the left and the right side of Eq. (83).

B. Inelastic contribution $\propto R^4$

We represent the total $E_1^{(6)}$ correction as a sum of four parts:

$$E_1^{(6)} = E_{L1} + E_Q + E'_Q + E_{H1}(p). \tag{88}$$

The middle energy contribution E_Q comes from momenta $q \sim \sqrt{2\,m\,\Lambda}$. We derive it by considering the nonrelativistic three Coulomb photon exchange,

$$E_Q = -\langle\phi,\phi_N|\frac{\alpha}{|\vec r - \vec R|}\,\frac{1}{E - H_0 + E_N - H_N}\,\frac{\alpha}{|\vec r - \vec R|}\,\frac{1}{E - H_0 + E_N - H_N}\,\frac{\alpha}{|\vec r - \vec R|}|\phi,\phi_N\rangle, \tag{89}$$

where H_0 is the lepton kinetic energy operator. In the corresponding electronic matrix element P_Q, one neglects the lepton binding energy E,

$$P_Q = -\phi^2(0)\,\alpha^3\,4\,m^2\,(4\,\pi)^3\int\frac{d^dq_1}{(2\,\pi)^d}\,\frac{d^dq_2}{(2\,\pi)^d}\,\frac{e^{i\,\vec q\,\vec R_1}}{q^2}\,\frac{1}{q^2 + 2\,m\,\Lambda}\,\frac{e^{-i\,(\vec q+\vec q\,')\vec R_2}}{(\vec q + \vec q\,')^2}\,\frac{1}{q'^2 + 2\,m\,\Lambda'}\,\frac{e^{i\,\vec q\,'\vec R_3}}{q'^2}, \tag{90}$$

expands in R_i, keeping terms $\propto R^4$, with the result

$$E_Q = -\phi^2(0)\,\alpha^3\,m^2\,\pi\left[\langle R^4\rangle\left(\frac{4}{15}\,\ln 2 - \frac{2}{5}\right) + \langle R^2\rangle^2\left(-\frac{10}{27} + \frac{2}{3}\,\ln 2 + \frac{2}{9}\,\ln\frac{\langle E\rangle_1}{m} - \frac{8}{9}\,\beta - \frac{1}{9\,\epsilon}\right)\right], \tag{91}$$

where

$$\ln\frac{\langle E\rangle_1}{m} = -\frac{1}{\langle R^2\rangle^2}\left[\langle 0|R^2\,\ln\frac{(H-E)'}{m}\,R^2|0\rangle - \frac{6}{5}\,\langle 0|R^i\,\ln\frac{(H-E)'}{m}\,R^2\,R^j|0\rangle\right.$$
$$\left. + \frac{3}{10}\,\langle 0|(R^i\,R^j - \delta^{ij}\,R^2/3)\,\ln\frac{(H-E)'}{m}\,(R^i\,R^j - \delta^{ij}\,R^2/3)|0\rangle\right]. \tag{92}$$

The average energy $\langle E\rangle_1$ does not depend on the lepton mass m, since the dependence on m cancels out between the left and right side of above equation. We calculate $\langle E\rangle_1$ by using the AV18 deuteron potential [19], with the result

$$\langle E\rangle_1 = 2.93(3)\ \mathrm{MeV}. \tag{93}$$

Equation (91) involves the dimensionless parameter β defined by

$$\beta = -\sum_{\Lambda_1,\Lambda_2}{}'\,[\langle 0|R^i\,R^j + 3\,\delta^{ij}\,R^2|\Lambda_1\rangle\,\langle\Lambda_1|R^i|\Lambda_2\rangle\,\langle\Lambda_2|R^j|0\rangle + \langle 0|R^i|\Lambda_1\rangle\,\langle\Lambda_1|\delta^{ij}\,R^2 - 3\,R^i\,R^j|\Lambda_2\rangle\,\langle\Lambda_2|R^j|0\rangle$$
$$+ \langle 0|R^i|\Lambda_1\rangle\,\langle\Lambda_1|R^j|\Lambda_2\rangle\,\langle\Lambda_2|\delta^{ij}\,R^2 - 3\,R^i\,R^j|0\rangle]\,\frac{3}{10\,\langle R^2\rangle^2}\,f\left(\frac{\Lambda_1}{\Lambda_2}\right), \tag{94}$$

with

$$f(x) = x\,\ln\left(1 + \frac{1}{\sqrt{x}}\right) - \sqrt{x} - \ln(1 + \sqrt{x}). \tag{95}$$

Since f weakly depends on its argument, one can replace the argument of f in Eq. (94) by its averaged value to obtain

$$\beta = f\left(\left\langle\frac{\Lambda_1}{\Lambda_2}\right\rangle\right). \tag{96}$$

For the estimation of β we will assume that $\langle\Lambda_1/\Lambda_2\rangle = 1,2,1/2$ and thus obtain $\beta = -1.0\,(0.2)$.

There is an additional contribution E'_Q that includes the finite proton size. It is obtained by inserting the proton electric formfactor in the Coulomb interaction in Eq. (89) and expanding in R_i up to the second order,

$$E'_Q = \phi^2(0)\,\alpha^3\,m^2\,r_s^2\,r_p^2\,\frac{4\,\pi}{9}\left[\frac{13}{3}+\frac{1}{2\,\epsilon}-5\,\ln 2-\frac{1}{r_s^2}\,\langle 0|\vec{R}\,\ln\frac{(H-E)}{m}\,\vec{R}|0\rangle\right]. \tag{97}$$

The remaining contributions E_{L1} is given by Eq. (47) with the deuteron radii, whereas $E_{H1}(p)$ is given by Eq. (63) with the proton charge radii.

Adding all parts together, the total nuclear structure contribution $E_1^{(6)} \propto R^4$ is

$$E_1^{(6)}(nS) = \alpha^6\,m^5\,\frac{4}{9\,n^3}\left\{r_d^4\left[-\frac{1}{n}+\gamma-\ln\frac{n}{2}+\Psi(n)+\ln\alpha\right]+r_{dd}^4\,\frac{3}{20\,n^2}-r_{ss}^4\left(\frac{3}{5}\,\ln 2-\frac{9}{10}\right)\right.$$
$$\left.+r_s^4\left(\frac{1}{3}-\frac{3}{2}\,\ln 2-\frac{1}{2}\,\ln\frac{\langle E\rangle_1}{m}+2\,\beta\right)+r_p^4\,[2+\gamma+\ln(r_{p1}\,m)]+r_s^2\,r_p^2\left(\frac{10}{3}-5\,\ln 2-\ln\frac{\langle E\rangle_2}{m}\right)\right\}. \tag{98}$$

Again, the part of the elastic contribution depending on the effective deuteron radius r_{d1} is not present in total $E_1^{(6)}$. The expression for the separate inelastic contribution is

$$E_{1,\rm pol}^{(6)} = E_1^{(6)} - E_{1,\rm fns}^{(6)} = \frac{\alpha^6}{n^3}\,m^5\,\delta_{l0}\left\{-r_{ss}^4\left(\frac{4}{15}\,\ln 2-\frac{2}{5}\right)+\frac{2}{9}\,r_s^4\left(\frac{2}{3}-3\,\ln 2-\ln\frac{\langle E\rangle_1}{m}+4\,\beta\right)\right.$$
$$\left.+\frac{4}{9}\,r_p^4\,[2+\gamma+\ln(r_{p1}\,m)]+\frac{4}{9}\,r_s^2\,r_p^2\left(\frac{10}{3}-5\,\ln 2-\ln\frac{\langle E\rangle_2}{m}\right)-\frac{4}{9}\,r_d^4\left[2+\gamma+\ln(r_{d1}\,m)\right]\right\}. \tag{99}$$

C. Total inelastic part

The sum $E_{1,\rm pol}^{(6)} + E_{2,\rm pol}^{(6)}$, as given by Eqs. (99) and (86), is the total three-photon exchange inelastic nuclear structure contribution, which is the main result of this work. It should be pointed out that several approximations have been made in our derivation of this result. First, we ignored the magnetic dipole and the electric quadrupole moments of deuteron. Second, we neglected the higher orders in $(H_N - E_N)/m$. These approximations contribute to the uncertainty of the inelastic part, which we estimate as 10%.

The remaining part of the total three-photon exchange nuclear structure contribution of Eq. (72) is the contribution due to the interaction with individual nucleons $E_{\rm np}^{(6)}$. We have little knowledge about the inelastic three-photon exchange between the muon and the proton but we expect it could be accounted for in terms of the same effective radii r_{p1} and r_{p2} as in the elastic part. We estimate the uncertainty associated with $E_{\rm np}^{(6)}$ in μH and in μD by applying Eq. (68) to the proton ($r_C \to r_p$) and making the following substitution,

$$\ln r_{p1} \to \ln r_{p1} \pm 1,$$
$$\ln r_{p2} \to \ln r_{p2} \pm 1. \tag{100}$$

It should be mentioned that $E_{\rm np}^{(6)}$ does not contribute to the μD-μH isotope shift difference.

VII. INELASTIC THREE-PHOTON EXCHANGE CORRECTION IN ORDINARY DEUTERIUM

The total inelastic nuclear structure α^6 correction for ordinary deuterium is split into four parts,

$$E^{(6)} = E_{L2} + E_R + E_{H2}(p) + E_{\rm np}^{(6)}, \tag{101}$$

where E_{L2} is given by Eq. (48) with the deuteron radii, and $E_{H2}(p)$ by Eq. (67) with the proton radii, while E_R is a Coulomb correction to the electric dipole polarizability, as given by Eq. (77), and $E_{\rm np}^{(6)}$ is the correction due to the interaction with individual nucleons.

Assuming that $H_N - E_N$ is much larger than the electron mass m, we obtain

$$E_R = -\pi\alpha^3\phi^2(0)\frac{1}{d}\langle\phi_N|R^k\left[\frac{1}{2\epsilon}+\frac{5}{2}-2\ln\frac{2(H_N-E_N)}{m}\right]$$
$$\times R^k|\phi_N\rangle + O\left(\frac{m}{H_N-E_N}\right). \tag{102}$$

We note that the neglected $O[m/(H_N - E_N)]$ terms do not vanish for the $l > 0$ states. Therefore, the nuclear polarizability correction does not vanish for the $l > 0$ states, but it is additionally suppressed by the ratio of the electron mass to the nuclear excitation energy.

The correction due to the interaction with individual nucleons $E_{\rm nucleon}^{(6)}$ is expected to be small and, moreover, it cancels out in the eD-eH isotope shift difference. To estimate the three-photon exchange of the bound electron with the proton, we use the same argumentation as in Ref. [14] to obtain

$$E_{\rm np}^{(6)}(p) = \frac{2\,\pi}{3}\,\alpha^3\,\phi^2(0)\,r_p^2\,\ln\frac{\bar{E}_p}{m} \tag{103}$$

and assume the uncertainty of 100%. The above correction is proportional to the squared charge radius, so the corresponding contribution for the neutron is negligible.

Our final result for the three-photon exchange nuclear structure correction in deuterium is given by

$$E^{(6)}(nS) = -\alpha^6\, m^3\, \frac{2}{3\,n^3}\left\{r_d^2\left[\frac{9}{4n^2} - 3 - \frac{1}{n} + \gamma - \ln\frac{n}{2} + \Psi(n) + \ln\alpha\right] + r_s^2\left(\frac{23}{12} - \ln\frac{2\,\langle E\rangle_2}{m}\right) + r_p^2\,[\gamma + \ln(r_{p2}\,m)]\right\} + E^{(6)}_{\rm np}. \tag{104}$$

Separately, the inelastic part $E^{(6)}_{\rm pol} = E^{(6)} - E^{(6)}_{\rm fns}$ is

$$E^{(6)}_{\rm pol} = -\frac{2\,\alpha^6}{3\,n^3}\,m^3\,\delta_{l0}\left\{r_s^2\left(\frac{23}{12} - \ln\frac{2\,\langle E\rangle_2}{m}\right) + r_p^2\,[\gamma + \ln(r_{p2}\,m)] - r_d^2\,[\gamma + \ln(r_{d2}\,m)]\right\} + E^{(6)}_{\rm np}. \tag{105}$$

We note that the fermion mass m cancels exactly in the expression in braces in the above equation.

VIII. RESULTS AND SUMMARY

Our numerical results for the three-photon exchange nuclear structure corrections are presented in Table II. The elastic part has been calculated with the exponential model of the nuclear charge distribution. It is displayed in the table separately for the comparison with the literature results. This part does not bear any uncertainty because its dependence on the charge distribution model cancels out exactly in the sum with the inelastic part. We observe a reasonable (although not perfect) agreement with the literature results summarized in Table II.

The inelastic three-photon exchange nuclear structure correction was calculated only for the electronic and muonic deuterium atoms; the corresponding results are presented in Table II. We find that the inelastic contribution for deuterium is of opposite sign as compared to its elastic counterpart and changes significantly the total $m\alpha^6$ nuclear structure contribution. In the case of eD, the change is of about 30%, while for μD, the inelastic part reverses the sign of the overall contribution. For electronic and muonic hydrogen, we present only estimations for the inelastic three-photon exchange contribution.

Our results for the three-photon exchange nuclear structure corrections affect determinations of the hydrogen-deuterium nuclear charge radii differences derived from the spectroscopic observations of the isotope shifts in electronic and muonic hydrogen and deuterium [2,20,21]. For the electronic H-D isotope shift of the $1S$-$2S$ transition, our result shifts the total theoretical prediction by 0.8 kHz, which is slightly larger than the theoretical error of 0.6 kHz assumed in Ref. [21]. There are, however, further corrections to the summary of theoretical contributions presented in Ref. [21], so we had to update it. Our review of the present status of the theory of the H-D isotope shift described in Appendix C leads us to the updated result for the nuclear charge radius difference determined from the measurement of the H-D isotope shift of the $1S$-$2S$ transition [20],

$$\delta r^2[\text{electronic}] \equiv r_d^2 - r_p^2 = 3.820\,70\,(31)\,\text{fm}^2, \tag{106}$$

which agrees with but is twice as accurate as the previous value of 3.820 07 (65) fm^2 obtained in Ref. [21].

For muonic hydrogen and deuterium, our result for the inelastic three-photon exchange nuclear structure contribution to the $2P_{1/2}$-$2S$ transition energy of 0.008 75 (88) meV shifts the deuteron-proton charge radius difference determined in Ref. [2] by 0.0014 fm^2, with the result

$$\delta r^2[\text{muonic}] \equiv r_d^2 - r_p^2 = 3.8126\,(34)\,\text{fm}^2. \tag{107}$$

The results derived from the electronic and muonic atoms disagree by about $2\,\sigma$, which confirms the discrepancy previously observed in Ref. [2].

In summary, we have studied the three-photon exchange $O(\alpha^2)$ nuclear structure correction to energy levels and the isotope shift of hydrogenlike muonic and electronic atoms. Our formula for the elastic contribution is valid for an arbitrary hydrogenic system and is much simpler than corresponding formulas in the literature [9]. The inelastic part has been derived for muonic and electronic deuterium only. Calculations of the three-photon inelastic contribution for He$^+$ and heavier elements are possible but are more complicated. At the same time, one may expect the inelastic contribution to be as large as the elastic part, which is a sizable correction in He$^+$, about 1% of the total nuclear nuclear size effect.

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APPENDIX A: DIMENSIONAL REGULARIZATION FOR BOUND STATES

The principles of dimensional regularization for bound states have been described in Ref. [26]. Here we only present formulas without derivation which have been used in the presented calculations. The dimension of space is assumed to be $d = 3 - 2\,\epsilon$. The surface area of the d-dimensional unit sphere is

$$\Omega_d = \frac{2\,\pi^{d/2}}{\Gamma(d/2)}. \tag{A1}$$

The Coulomb potential in d dimensions is of the form

$$\mathcal{V}(r) = \int \frac{d^d k}{(2\,\pi)^d}\,\frac{4\,\pi}{k^2}\,e^{i\,\vec{k}\cdot\vec{r}} = \frac{C_1}{r^{1-2\,\epsilon}}, \tag{A2}$$

where

$$C_1 = \pi^{\epsilon-1/2}\,\Gamma(1/2-\epsilon). \tag{A3}$$

The elastic contribution involves another potential of the form

$$\mathcal{V}^{(2)}(r) = \int \frac{d^d k}{(2\,\pi)^d}\,\frac{4\,\pi}{k^4}\,e^{i\,\vec{k}\cdot\vec{r}} = C_2\,r^{1+2\,\epsilon}, \tag{A4}$$

TABLE II. Numerical results for the three-photon exchange nuclear structure corrections. Numerical values include the leading recoil effect by the multiplicative reduced-mass prefactor $(\mu/m)^3$. Elastic contributions are obtained with the exponential parametrization of the nuclear charge distribution, with the following values of nuclear radii: $r_p = 0.84087$ fm, $r_d = 2.12562$ fm, $r_C(^3\text{He}) \equiv r_h = 1.973$ fm [22], $r_C(^4\text{He}) \equiv r_\alpha = 1.681$ fm [23].

Transition	Units	Elastic	Inelastic	Sum	Elastic by others
$E^{(6)}(2S{-}1S,e\text{H})$	Hz	−584	−344 (344)	−928 (344)	−587 (2)[a]
$E^{(6)}(2S{-}1S,e\text{D}{-}e\text{H})$	Hz	−2 846	817 (41)	−2 029 (41)	−2 834 (13)[a]
$E^{(6)}(2P_{1/2}{-}2S,\mu\text{H})$	meV	−0.001 27	±0.000 27	−0.001 27 (27)	−0.001 34[b]
$E^{(6)}(2P_{1/2}{-}2S,\mu\text{D})$	meV	−0.006 56	0.008 75 (88)(27)[f]	0.002 19 (88)(27)[f]	−0.006 50 (60)[c]
$E^{(6)}(2P_{1/2}{-}2S,\mu^3\text{He}^+)$	meV	−0.384 7	unknown		−0.378 6 (60)[d]
$E^{(6)}(2P_{1/2}{-}2S,\mu^4\text{He}^+)$	meV	−0.304 8	unknown		−0.311 5 (140)[e]

[a]CODATA [16].
[b]Reference [11], the difference of entries "Our choice" and "Non-rel. finite-size" in Table 2 of that work, $-0.0019\, r_p^2$.
[c]Reference [12], the sum of entries r_3 and r_3' in Table 2 of that work, $-0.002\,124\,(4)\, r_d^2 + 0.003\,10\,(60)$ meV.
[d]Reference [24], the sum of entries r_3 and r_3' in Table 2 of that work, $-0.1288\,(13)\, r_h^2 + 0.1177\,(33)$ meV.
[e]Reference [25], the sum of entries r_3 and r_3' in Table 4 of that work, $-0.1340\,(30)\, r_\alpha^2 + 0.0672\,(112)$ meV.
[f]The second uncertainty comes from the interaction with individual nucleons and cancels in the μD-μH isotope shift.

where

$$C_2 = \tfrac{1}{4}\,\pi^{\epsilon-1/2}\,\Gamma(-1/2-\epsilon). \tag{A5}$$

Furthermore, we used the following integration formulas:

$$\int_\Lambda d^d r\,[\mathcal{V}(r)]^3 = -[(4\,\pi)^\epsilon\,\Gamma(1+\epsilon)]^2\,4\,\pi\,\left[\frac{1}{4\,\epsilon} + \frac{1}{2} + \gamma + \ln(\Lambda)\right], \tag{A6}$$

$$\int \frac{d^d k}{(2\pi)^d}\,\frac{1}{k^{2\alpha}}\,\frac{1}{(k-q)^{2\beta}} = \frac{[q^2]^{\frac{d}{2}-\alpha-\beta}}{[4\pi]^{\frac{d}{2}}}\,\frac{\Gamma\big(\alpha+\beta-\frac{d}{2}\big)\,\Gamma\big(\frac{d}{2}-\alpha\big)\,\Gamma\big(\frac{d}{2}-\beta\big)}{\Gamma(d-\alpha-\beta)\,\Gamma(\alpha)\,\Gamma(\beta)}, \tag{A7}$$

and [31]

$$\begin{aligned} I &= \int \frac{d^d k}{(2\,\pi)^d} \int \frac{d^d q}{(2\,\pi)^d}\,\frac{1}{[k^2]^{n_1}}\,\frac{1}{\big[(k-q)^2+m_2^2\big]^{n_2}}\,\frac{1}{\big[q^2+m_3^2\big]^{n_3}} \\ &= \frac{m_3^{2\,(d-n_1-n_2-n_3)}}{(4\,\pi)^d}\,\frac{\Gamma(d/2-n_1)\,\Gamma(n_1+n_2-d/2)\,\Gamma(n_1+n_3-d/2)\,\Gamma(n_1+n_2+n_3-d)}{\Gamma(2\,n_1+n_2+n_3-d)\,\Gamma(n_2)\,\Gamma(n_3)\,\Gamma(d/2)} \\ &\quad\times {}_2F_1\big(n_1+n_2+n_3-d, n_1+n_2-d/2, 2\,n_1+n_2+n_3-d, 1-m_2^2/m_3^2\big). \end{aligned} \tag{A8}$$

APPENDIX B: NUMERICAL VERIFICATION OF THE ELASTIC CONTRIBUTION

The finite nuclear size (fns) correction can be calculated numerically to all orders in $Z\alpha$, by computing the energy eigenvalue of the Dirac equation with the extended-size nuclear potential and subtracting the analytical point-nucleus result. Knowing the leading α^4 and α^5 fns corrections analytically, we can also identify the higher-order fns residual from the numerical all-order results.

The main problem in determining the fns correction numerically is that the corresponding effect is very small for light electronic atoms. So, for the $2s$ state of hydrogen, the relativistic $O(\alpha^2)$ fns correction yields a 1×10^{-13} fraction of the binding energy. To make an extensive comparison between the numerical and analytical approaches, we performed numerical calculations for Z as low as $Z = 0.25$. To make sure that possible numerical uncertainties do not interfere with the comparison, we determined the binding energies with a 20-digit numerical precision.

To compute the eigenvalues of the Dirac equation, we use the Dual Kinetic Balance method [27] with the finite basis set of B-splines. Because of high accuracy demands, we implemented this method in quadruple (about 32 digits) arithmetics, similarly as it was done recently in calculations of the recoil corrections [28]. About 200–250 basis functions were sufficient to reach the required 20-digit numerical accuracy for the binding energies.

We obtained the relativistic fns correction $E^{(6+)}_{\text{fns}}$ that contains contributions of order $(Z\alpha)^6$ and higher as

$$E^{(6+)}_{\text{fns}} = E_{\text{fns}} - E^{(4)}_{\text{fns}} - E^{(5)}_{\text{fns}}, \tag{B1}$$

where E_{fns} is determined numerically by solving the Dirac equation, $E^{(4)}_{\text{fns}}$ is given by Eq. (6), $E^{(5)}_{\text{fns}}$ is given by Eq. (14), and r_Z is evaluated for the same nuclear model as in the numerical calculation.

The comparison of our all-order numerical results for $E^{(6+)}_{\text{fns}}$ with the analytical $(Z\alpha)^6$ result $E^{(6)}$ given by Eq. (68) is presented in Fig. 1. Both numerical and analytical results are

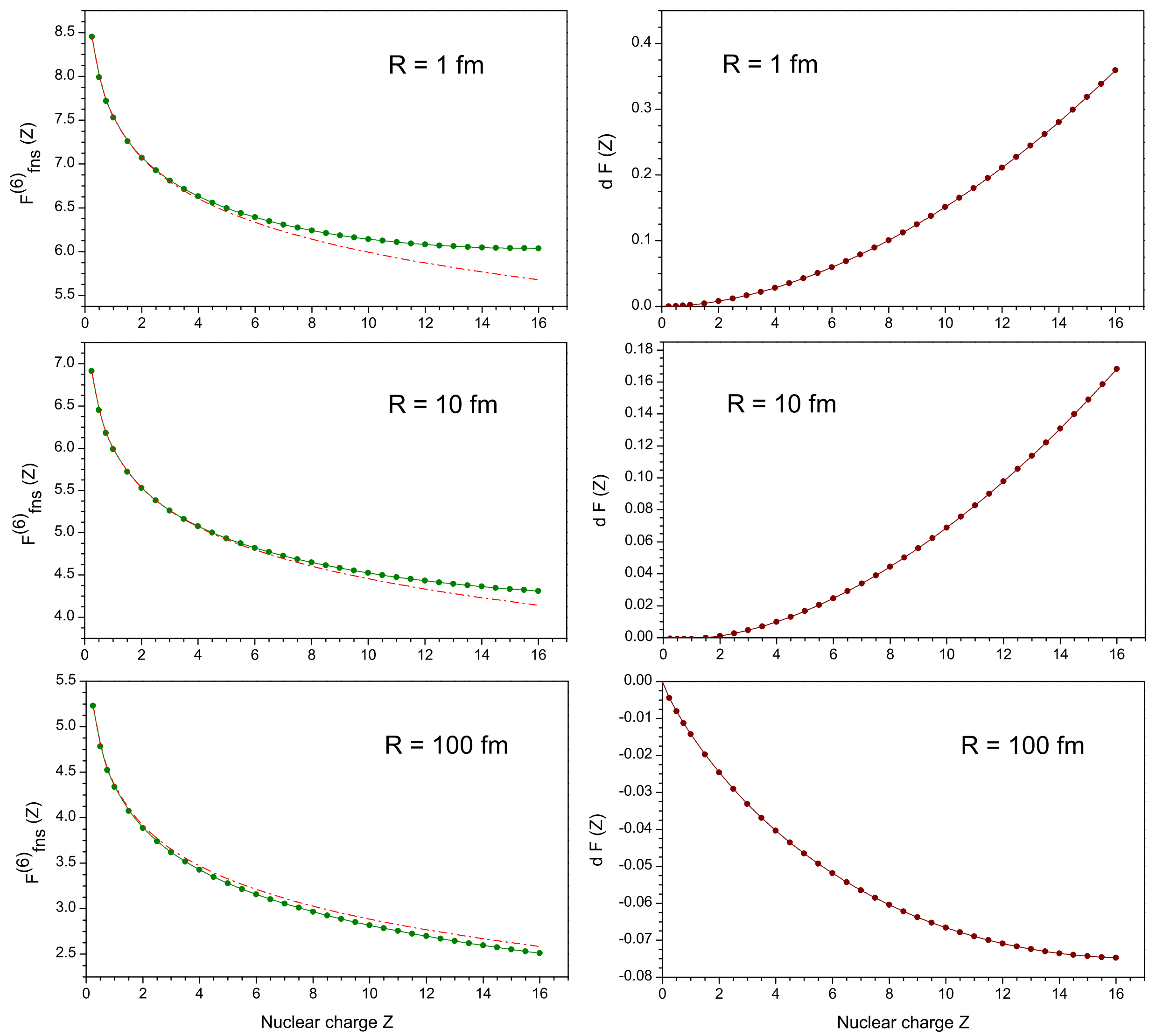

FIG. 1. The relativistic finite nuclear size correction for the hydrogenic 1s state is plotted as a function of Z for three different nuclear radii: $r_C = 1$ fm (upper row), $r_C = 10$ fm (middle row), and $r_C = 100$ fm (lower row). In each row, the left graph shows a comparison of the numerical function $F^{(6+)}_{\rm fns}$ (filled dots and solid line, green) with the analytical function $F^{(6)}_{\rm fns}$ (dash-dotted line, red); the right graphs show the remainder function $\delta F = F^{(6+)}_{\rm fns} - F^{(6)}_{\rm fns}$.

obtained with the exponential model of the nuclear charge distribution (see Table I). We plot the scaled function with the leading $Z\alpha$, r_C, and n dependence removed,

$$F^{(6+)}_{\rm fns} = \frac{E^{(6+)}_{\rm fns}}{m^3 (Z\alpha)^6\, r_C^2/n^3}. \tag{B2}$$

As can be seen from the figure, agreement between the numerical and analytical results is excellent.

APPENDIX C: HYDROGEN-DEUTERIUM 1S-2S ISOTOPE SHIFT

In this section we update the summary of all available theoretical contributions for the eH-eD isotope shift of the 1S-2S transition frequency reviewed previously by Jentschura *et al.* [21]. We use the following values for the fundamental constants, the fine-structure constant,

$$\alpha^{-1} = 137.035\,999\,139\,(31),$$

and the Rydberg constant,

$$R_\infty c = 3.289\,841\,960\,355\,(19) \times 10^{15}\,\text{Hz},$$

from CODATA 2014 [16]. The electron-proton mass ratio we take from the recent measurement by Heiße *et al.* [29],

$$\frac{m_p}{m_e} = 1\,836.152\,673\,346\,(81).$$

We note that this value is twice as accurate but 3 σ off from the CODATA 2014 value [16]. For the electron-deutron mass ratio we use the CODATA value [16],

$$\frac{m_D}{m_e} = 3\,670.482\,967\,85\,(13).$$

In the present section we follow the notations and conventions of Ref. [21]. We will not repeat the full review of the theory but only indicate the entries therein that need to be updated. The changes are as follows.

(i) The updated result for the leading (Dirac) contribution to the isotope shift (Eq. (28) of Ref. [21]) is

$$\Delta f_{\rm i} = 671\,004\,071.107\,(64)\ \text{kHz}, \tag{C1}$$

the change being due to the updated values of the electron-nucleus mass ratios.

(ii) Our present result for the two-photon exchange nuclear structure correction specified by Eqs. (32) and (40),

$$E^{(5)}(\text{H–D}, 1S\text{–}2S) = 19.12\,(20)\ \text{kHz}, \tag{C2}$$

replaces the sum of $\Delta\nu_9$ given by Eq. (40) of Ref. [21] and $E_{\rm NS,(b)}$ given by Eq. (45) therein, amounting to 19.11 (2) kHz.

(iii) Our present result for the three-photon exchange nuclear structure correction,

$$E^{(6)}(\text{H–D}, 1S\text{–}2S) = -2.029\,(41)\ \text{kHz}, \tag{C3}$$

replaces the sum of $E_{\rm NS,(c)}$ given by Eq. (47) of Ref. [21] and $\Delta\nu_{11} = \pm 0.5$ kHz given by Eq. (43) therein, amounting to -2.828 ± 0.5 kHz.

(iv) The entry for the higher-order pure recoil $\nu_5 = -3.41\,(32)$ kHz (Eq. (33) of Ref. [21]) is replaced by the complete all-order (in $Z\alpha$) result by Yerokhin and Shabaev [28,30]. The corresponding correction to the energy is

$$\delta E(nS) = \frac{m^2}{M}\frac{(Z\alpha)^5}{\pi n^3}\left[Z\alpha\left(4\ln 2 - \frac{7}{2}\right)\pi + (Z\alpha)^2\, G_{\rm rec} + \delta_{\rm fns} P\right], \tag{C4}$$

where $G_{\rm rec}(1S, Z=1) = 9.720\,(3)$, $G_{\rm rec}(2S, Z=1) = 14.899\,(3)$, $\delta_{\rm fns} P(nS,\text{H}) = -0.000\,184\,(1)$ in the case of hydrogen and $\delta_{\rm fns} P(nS,\text{D}) = -0.000\,786\,(6)$ for deuteron [28,30]. In the result, the updated contribution is

$$\Delta\nu_5 = -3.058\ \text{kHz}. \tag{C5}$$

(v) For the radiative recoil contribution (Eq. (36) of Ref. [21]), we use the estimation of uncertainty from Ref. [16], which is about three times larger than the one of Ref. [21],

$$\Delta\nu_6 = -5.38\,(35)\ \text{kHz}. \tag{C6}$$

The final theoretical value of

$$\Delta f_{\rm th} = 670\,999\,567.88\,(42)\ \text{kHz}, \tag{C7}$$

replaces the previous result $\Delta f_{\rm th}([21]) = 670\,999\,566.90\,(89)$ kHz. Combining the theoretical value $\Delta f_{\rm th}$ with the experimental result from Refs. [20,21], we obtain the updated result for the mean-square charge-radii difference,

$$r_d^2 - r_p^2 = 3.820\,70\,(31)\ \text{fm}^2, \tag{C8}$$

which agrees with but is twice as accurate as the previous value of 3.820 07 (65) fm^2 [21].

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