Editors' Suggestion

Relativistic corrections to the Bethe logarithm for the $2\,^3S$ and $2\,^3P$ states of He

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With this work we start a project calculating the QED contribution of order $\alpha^7 m$ to the $2\,^3P$–$2\,^3S$ transition energy in helium, aiming for an accurate determination of the nuclear charge radius r_E from measurements of the corresponding transition frequency. Together with the complementary determination of r_E from muonic helium, this project will provide a stringent test of universality of electromagnetic interactions of leptons in the Standard Model. We report a calculation of the relativistic corrections to the Bethe logarithm for the $2\,^3S$ and $2\,^3P$ states, which is the most numerically demanding part of the project.

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I. INTRODUCTION

One of the prominent low-energy tests of the Standard Model (SM) with a possible signature of new physics is based on a comparison of the Lamb shift in muonic hydrogen μH and electronic hydrogen H. The lepton universality of SM implies that the same physical laws and physical constants define the energy levels in H and μH. However, it has been found that the proton root-mean-square charge radius, extracted from the comparison of theory and experiment for the Lamb shift, turned out to be significantly different for the electronic [1] and muonic [2,3] hydrogen,

$$r_p(\mathrm{H}) = 0.875\,9\,(77)\ \mathrm{fm}\,,$$
$$r_p(\mu\mathrm{H}) = 0.840\,87\,(39)\ \mathrm{fm}.$$

This $4.5\,\sigma$ discrepancy, known as the proton radius puzzle, may signal the existence of interactions that are not accounted for in the Standard Model. Several experiments aiming to resolve the puzzle have been accomplished recently, namely, the measurement of the $2S$–$4P$ transition energy in Garching [4], $2S$–$2P$ Lamb shift in Toronto [5], and two measurements of the $1S$–$3S$ transition energy performed in Paris [6] and in Garching [7]. These experiments yield conflicting results for the proton charge radius, which does not solve the puzzle but suggests the presence of unknown systematic effects in hydrogen measurements. Further experiments directed to clarify the proton radius puzzle are currently being pursued, notably, measurements of the $1S$–$2S$ transition energy in He^+ [8,9], transitions between circular Rydberg states in H-like ions [10], and the direct comparison of the cross sections of the e-p versus μ-p elastic scattering [11].

An alternative way to solve the proton charge radius puzzle can be gained through spectroscopy of the helium atom. Specifically, a comparison of the nuclear charge radius from the helium spectroscopy with the radius from muonic helium, expected soon from the CREMA collaboration [12], would provide an independent test of the lepton universality in atomic systems. On the experimental side, several transition energies in the helium atom are already known with an accuracy sufficient for determining the nuclear charge radius on a 10^{-3} fractional level [13–19]. In order to achieve a similar level of accuracy in theoretical predictions, one needs to improve the previous helium calculations [20,21] by completing the next-order term of the NRQED expansion, namely, the $\alpha^7 m$ correction. This is a very challenging theoretical problem. Among few-electron atoms, it has so far been solved only for the helium fine structure [22–24].

With this paper, we start a project calculating the complete $\alpha^7 m$ correction for energy levels of two-electron atoms. At present, we restrict ourselves to the triplet states, for which the nonrelativistic wave function $\phi(\vec{r}_1,\vec{r}_2)$ vanishes at $\vec{r}_1 = \vec{r}_2$. As a result, the whole class of the so-called contact operators does not contribute, thus making the derivation of $\alpha^7 m$ operators more tractable. An improved theory of the triplet states will allow the determination of the nuclear charge radius from the 2^3S–2^3P transition in ^{4}He, which was accurately measured by the Hefei group [18].

The present status of theory of helium energy levels complete up to order $\alpha^6 m$ is described in our recent review [25]. The next-order $\alpha^7 m$ contribution can be represented as a sum of three parts,

$$E^{(7)} = \langle H^{(7)}\rangle + 2\left\langle H^{(4)}\,\frac{1}{(E-H)'}\,H^{(5)}\right\rangle + E_L\,, \tag{1}$$

where $H^{(4)}$, $H^{(5)}$, and $H^{(7)}$ are the effective Hamiltonians of order $\alpha^4 m$, $\alpha^5 m$, and $\alpha^7 m$, respectively; H and E are the nonrelativistic Hamiltonian and its eigenvalue, respectively; and E_L is the low-energy contribution, also known as the relativistic correction to the Bethe logarithm, which is the main subject of this work.

Out of the three terms contributing to $E^{(7)}$, the relativistic correction to the Bethe logarithm is numerically the most demanding one and thus is the crucial part of the whole $\alpha^7 m$ project. The calculation of (the spin-dependent part of) such a correction was first performed in Ref. [26] for the fine structure of helium and later improved in Refs. [23] and [24]. For the Coulomb two-center systems (H_2^+, HD^+, etc.), the relativistic corrections to the Bethe logarithm were calculated by Korobov *et al.* [27]. The goal of the present work is to calculate the spin-independent low-energy correction E_L for the $2\,^3S$ and $2\,^3P$ states of helium.

II. NONRELATIVISTIC LOW-ENERGY CONTRIBUTION

The leading nonrelativistic (dipole) low-energy contribution of order $\alpha^5\, m$ is given by

$$E_{L0}(\Lambda) = \frac{e^2}{m^2}\int_{k<\Lambda}\frac{d^3k}{(2\,\pi)^3\,2\,k}\left(\delta^{ij}-\frac{k^i\,k^j}{k^2}\right) \times\left\langle P^i\,\frac{1}{E-H-k}\,P^j\right\rangle, \tag{2}$$

where $\vec{P}=\vec{p}_1+\vec{p}_2$, and Λ is the high-momentum cutoff. $E_{L0}(\Lambda)$ diverges when $\Lambda\to\infty$ and requires subtraction of the leading terms in the large-Λ asymptotics.

Performing the angular integration and dropping the overall prefactor $\alpha^5 m$, one obtains

$$E_{L0}(\Lambda) = \frac{2}{3\,\pi}\int_0^\Lambda k\,dk\,P(k)\,, \tag{3}$$

where

$$P(k) = \left\langle \vec{P}\,\frac{1}{E-H-k}\,\vec{P}\right\rangle. \tag{4}$$

The large-k expansion of $P(k)$ is

$$k\,P(k) = -\langle P^2\rangle + \frac{D}{k}+\cdots\,, \tag{5}$$

where $D=2\,\pi\,Z\,\langle\delta^3(r_1)+\delta^3(r_2)\rangle$. The finite part of the low-energy contribution is then defined by dropping terms proportional to Λ and $\ln(2\,\Lambda)$ as

$$E_{L0} = -\frac{2}{3\,\pi}\,D\,\ln k_0\,, \tag{6}$$

where $\ln k_0$ is the standard Bethe logarithm,

$$\ln k_0 = -\frac{1}{D}\int_0^\infty dk\left[k\,P(k)+\langle P^2\rangle-\frac{D}{k}\,\theta(2k-1)\right] = \frac{\langle\vec{P}\,(H-E)\,\ln[2\,(H-E)]\,\vec{P}\rangle}{2\,\pi\,Z\left\langle\sum_a\delta^3(r_a)\right\rangle}\,, \tag{7}$$

and $\theta(x)$ is the Heaviside step function.

The numerical calculation of the Bethe logarithm for the helium atom remained for a long time a very difficult problem [28,29], which has been successfully solved only relatively recently [30–32].

III. RELATIVISTIC LOW-ENERGY CORRECTIONS

There are three types of relativistic corrections of order $\alpha^7 m$ to the low-energy contribution (2),

$$E_L = E_{L1}+E_{L2}+E_{L3}\,. \tag{8}$$

The first part E_{L1} is a perturbation of the nonrelativistic low-energy contribution E_{L0} in Eq. (2) by the Breit Hamiltonian $H^{(4)}$, the second part E_{L2} is induced by the relativistic correction to the current operator $\vec{P}$, whereas the third term E_{L3} is the retardation correction. All of these corrections are defined as remainders after dropping divergent in Λ terms, such as Λ and $\ln\Lambda$.

A. Breit correction E_{L1}

The low-energy contribution perturbed by the (spin-independent part of the) Breit Hamiltonian $H^{(4)}$ is

$$E_{L1}(\Lambda) = \frac{2}{3\,\pi}\int_0^\Lambda k dk\,P_{L1}(k)\,, \tag{9}$$

where

$$P_{L1}(k) = 2\left\langle H^{(4)}\frac{1}{(E-H)'}\,\vec{P}\,\frac{1}{E-H-k}\,\vec{P}\right\rangle + \left\langle\vec{P}\,\frac{1}{E-H-k}\big[H^{(4)}-\langle H^{(4)}\rangle\big]\frac{1}{E-H-k}\,\vec{P}\right\rangle, \tag{10}$$

where (with $r\equiv r_{12}$)

$$H^{(4)} = -\frac{1}{8}\left(p_1^4+p_2^4\right)+\frac{Z\,\pi}{2}[\delta^3(r_1)+\delta^3(r_2)]+\pi\,\delta^3(r) - \frac{1}{2}\,p_1^i\left(\frac{\delta^{ij}}{r}+\frac{r^i\,r^j}{r^3}\right)p_2^j. \tag{11}$$

The large-k expansion of $P_{L1}(k)$ is given by

$$k\,P_{L1}(k) = A_1+\frac{B_1}{\sqrt{k}}+\frac{C_1\,\ln k}{k}+\frac{D_1}{k}+\cdots\,, \tag{12}$$

where the asymptotic constants are derived in Appendix A.

We construct the finite part of the Breit correction as

$$E_{L1} = \frac{2}{3\,\pi}\int_0^\infty dk\left[k\,P_{L1}(k)-A_1 - \frac{B_1}{\sqrt{k}}-\left(\frac{C_1\,\ln k}{k}+\frac{D_1}{k}\right)\theta(k-1)\right] \tag{13}$$

$$= \frac{2}{3\pi}\left\{\int_0^K kdk\,P_{L1}(k)+\int_K^\infty dk\left[k\,P_{L1}(k)-A_1 - \frac{B_1}{\sqrt{k}}-\frac{C_1\ln k}{k}-\frac{D_1}{k}\right] - \left[A_1\,K+2\,B_1\sqrt{K}+\frac{C_1}{2}\ln^2 K+D_1\,\ln K\right]\right\}, \tag{14}$$

where $K\geqslant 1$ is a free parameter.

B. Current correction E_{L2}

The second low-energy contribution of order $\alpha^7 m$ is induced by a correction to the current operator in Eq. (2), $\vec{P}\to\vec{P}+\delta\,\vec{j}$, with

$$\delta j^i = i\big[H^{(4)},r_1^i+r_2^i\big] = -\frac{1}{2}\left(p_1^i\,p_1^2+p_2^i\,p_2^2\right)-\frac{1}{2}\left(\frac{\delta^{ij}}{r}+\frac{r^i r^j}{r^3}\right)\left(p_1^j+p_2^j\right), \tag{15}$$

where [,] denotes a commutator. The corresponding low-energy correction is

$$E_{L2}(\Lambda) = \frac{4}{3\pi}\int_0^\Lambda k dk\, P_{L2}(k)\,, \tag{16}$$

where

$$P_{L2}(k) = \left\langle \vec{\delta j}\, \frac{1}{E-H-k}\, \vec{P} \right\rangle. \tag{17}$$

The large-k expansion of $P_{L2}(k)$ has the same form as that of $P_{L1}(k)$,

$$k\, P_{L2}(k) = A_2 + \frac{B_2}{\sqrt{k}} + \frac{C_2\,\ln k}{k} + \frac{D_2}{k} + \cdots\,, \tag{18}$$

where the asymptotic constants are derived in Appendix B.

The finite part of the low-energy correction is constructed as

$$E_{L2} = \frac{4}{3\pi}\int_0^\infty dk\left[k\, P_{L2}(k) - A_2 - \frac{B_2}{\sqrt{k}} - \left(\frac{C_2\,\ln k}{k} + \frac{D_2}{k}\right)\theta(k-1)\right] \tag{19}$$

$$= \frac{4}{3\pi}\left\{\int_0^K k dk\, P_{L2}(k) + \int_K^\infty k dk\left[P_{L2}(k) - \frac{A_2}{k} - \frac{B_2}{k^{3/2}} - \frac{C_2\ln k}{k^2} - \frac{D_2}{k^2}\right] - \left[A_2\,K + 2B_2\sqrt{K} + \frac{C_2}{2}\ln^2 K + D_2\ln K\right]\right\}, \tag{20}$$

where $K \geqslant 1$ is a free parameter.

C. Retardation correction E_{L3}

A retardation correction to the low-energy contribution is

$$E_{L3}(\Lambda) = \frac{2}{3\pi}\int_0^\Lambda k dk\, P_{L3}(k)\,, \tag{21}$$

where

$$P_{L3}(k) = \frac{3}{8\pi}\int d\Omega_k\left(\delta^{ij} - \frac{k^i\,k^j}{k^2}\right)\sum_{a,b=1,2}\delta_{k^2}\left\langle p_a^i\, e^{i\,\vec{k}\cdot\vec{r}_a}\, \frac{1}{E-H-k}\, p_b^j\, e^{-i\,\vec{k}\cdot\vec{r}_b}\right\rangle, \tag{22}$$

where $\delta_{k^2}\langle\ldots\rangle$ denotes the quadratic in k term of the small-k expansion of the exponential functions in the matrix element $\langle\ldots\rangle$. Performing the expansion and integrating over angular variables, we obtain

$$P_{L3}(k) = \frac{3}{8\pi}\int \Omega_k\left(\delta^{ij} - \frac{k^i\,k^j}{k^2}\right)\sum_{a,b=1,2}\left\langle p_a^i\,(\vec{k}\cdot\vec{r}_a)\,\frac{1}{E-H-k}\,(\vec{k}\cdot\vec{r}_b)\,p_b^j - p_a^i\,(\vec{k}\cdot\vec{r}_a)^2\,\frac{1}{E-H-k}\,p_b^j\right\rangle \tag{23}$$

$$= \frac{k^2}{10}\left[3\left\langle\left(p_1^i r_1^j + p_2^i r_2^j\right)^{(2)}\frac{1}{E-H-k}\left(r_1^j p_1^i + r_2^j p_2^i\right)^{(2)}\right\rangle - \frac{5}{2\,k}\,\langle\vec{L}^2\rangle\right.$$
$$\left. - 2\left\langle\left[p_1^i\left(2\,\delta^{ij} r_1^2 - r_1^i r_1^j\right) + p_2^i\left(2\,\delta^{ij} r_2^2 - r_2^i r_2^j\right)\right]\frac{1}{E-H-k}\left(p_1^j + p_2^j\right)\right\rangle\right], \tag{24}$$

where $(a^i b^j)^{(2)} = (a^i b^j + a^j b^i)/2 - \vec{a}\cdot\vec{b}\,\delta^{ij}/3$ and $\vec{L} = \vec{r}_1\times\vec{p}_1 + \vec{r}_2\times\vec{p}_2$. The large-$k$ expansion of $P_{L3}(k)$ is of the form

$$k\, P_{L3}(k) = G_3\,k^2 + F_3\,k + A_3 + \frac{B_3}{\sqrt{k}} + \frac{C_3\,\ln k}{k} + \frac{D_3}{k} + \cdots\,, \tag{25}$$

where the asymptotic constants are derived in Appendix C.

We construct the finite part of the retardation correction as

$$E_{L3} = \frac{2}{3\pi}\int_0^\infty dk\left\{k\, P_{L3}(k) - k^2\,G_3 - k\,F_3 - A_3 - \frac{B_3}{\sqrt{k}} - \left[\frac{C_3\,\ln k}{k} + \frac{D_3}{k}\right]\theta(k-1)\right\} \tag{26}$$

$$= \frac{2}{3\pi}\left\{\int_0^K dk\,k P_{L3}(k) + \int_K^\infty dk\left[k P_{L3}(k) - G_3 k^2 - F_3 k - A_3 - \frac{B_3}{\sqrt{k}} - \frac{C_3\ln k}{k} - \frac{D_3}{k}\right]\right.$$
$$\left. - \left[G_3\frac{K^3}{3} + F_3\frac{K^2}{2} + A_3\,K + 2\,B_3\sqrt{K} + \frac{C_3}{2}\ln^2 K + D_3\ln K\right]\right\}, \tag{27}$$

where $K \geqslant 1$ is a free parameter.

IV. NUMERICAL EVALUATION

A. Transformation to a regularized form

The Breit Hamiltonian $H^{(4)}$ [Eq. (11)] contains singular operators ($\delta(r_a)$, p_a^4) which complicates numerical evaluations of the Breit correction E_{L1}. In order to achieve high numerical accuracy, we transform Eq. (10) to a more regular form. Specifically, by using the identity

$$H^{(4)}|\phi\rangle = H_A^{(4)}|\phi\rangle + \{H - E, Q\}|\phi\rangle\,, \tag{28}$$

where $|\phi\rangle$ is the eigenfunction H with energy E,

$$Q = -\frac{1}{4}\left(\frac{Z}{r_1} + \frac{Z}{r_2} - \frac{2}{r}\right) \tag{29}$$

and

$$H_A^{(4)}|\phi\rangle = \left[-\frac{1}{2}(E - V)^2 + \frac{1}{4}\nabla_1^2\nabla_2^2 - \frac{Z}{4}\frac{\vec{r}_1}{r_1^3}\cdot\vec{\nabla}_1 - \frac{Z}{4}\frac{\vec{r}_2}{r_2^3}\cdot\vec{\nabla}_2 - \frac{1}{2}\, p_1^i\left(\frac{\delta^{ij}}{r} + \frac{r^i\, r^j}{r^3}\right) p_2^j\right]|\phi\rangle\,, \tag{30}$$

with $V = -Z/r_1 - Z/r_2 + 1/r$, we transform the first term in the right-hand side of Eq. (10) to a regularized form,

$$P_{\rm pwf}(k) = 2\left\langle H_A^{(4)} \frac{1}{(E - H)'}\,\vec{P}\,\frac{1}{E - H - k}\,\vec{P}\right\rangle - 2\left\langle[Q - \langle Q\rangle]\,\vec{P}\,\frac{1}{E - H - k}\,\vec{P}\right\rangle. \tag{31}$$

Furthermore, by using the identity

$$H^{(4)} = H_B^{(4)} + \{H - E, Q_B\} - \frac{1}{2}(H - E)^2\,, \tag{32}$$

where

$$Q_B = -\frac{E}{2} - \frac{1}{4}\left(\frac{Z}{r_1} + \frac{Z}{r_2} - \frac{2}{r}\right), \tag{33}$$

and

$$H_B^{(4)} = -\frac{1}{2}(E - V)\left(E - \frac{1}{r}\right) + \frac{1}{4}\nabla_1^2\nabla_2^2 - \frac{Z}{4}\vec{p}_1\left(\frac{1}{r_1} + \frac{1}{r_2}\right)\vec{p}_1 - \frac{Z}{4}\vec{p}_2\left(\frac{1}{r_1} + \frac{1}{r_2}\right)\vec{p}_2 - \frac{1}{2}\, p_1^i\left(\frac{\delta^{ij}}{r} + \frac{r^i\, r^j}{r^3}\right) p_2^j\,, \tag{34}$$

we transform the second term in the right-hand side of Eq. (10) to the regularized form,

$$P_{\rm ver}(k) = \left\langle\vec{P}\,\frac{1}{E - H - k}\left[H_B^{(4)} - 2kQ_B - \frac{k^2}{2} - \langle H_B^{(4)}\rangle\right]\frac{1}{E - H - k}\,\vec{P}\right\rangle - \left\langle[2\vec{P}\,Q_B + k\vec{P}]\frac{1}{E - H - k}\,\vec{P}\right\rangle - \frac{1}{2}\langle\vec{P}^2\rangle\,. \tag{35}$$

B. Angular decomposition

In our approach, we express all wave functions and perform the angular momentum algebra in Cartesian coordinates. The reference-state wave functions of the ^3S and $^3P^o$ symmetry are represented as

$$\phi(^3S) = F(r_1, r_2, r) - (1 \leftrightarrow 2)\,, \tag{36}$$

$$\phi^i(^3P^o) = r_1^i\, F(r_1, r_2, r) - (1 \leftrightarrow 2)\,, \tag{37}$$

where the scalar functions F are linear combinations of exponential functions,

$$F(r_1, r_2, r) = \sum_i c_i \exp\left(-\alpha_i r_1 - \beta_i r_2 - \gamma_i r\right). \tag{38}$$

The wave functions are normalized by $\langle\phi|\phi\rangle = 1$ and $\langle\phi^i|\phi^i\rangle = 1$.

The angular decomposition of formulas in Sec. III is mostly performed in the same way as for the nonrelativistic Bethe logarithm. In that case, for the ^3S reference state, only $^3P^o$ intermediate states contribute,

$$\begin{aligned} P(k) &= \langle\phi(^3S)|\, P^i\left(\frac{1}{E - H - k}\right)_{^3P^o} P^i\,|\phi(^3S)\rangle \\ &= \langle\phi(^3S)|\, P^i\,|\,\delta\phi^i(^3P^o)\rangle\,, \end{aligned} \tag{39}$$

where $\delta\phi^i$ is the perturbed wave function of the $^3P^o$ symmetry,

$$\delta\phi^i(^3P^o) = \sum_n \frac{\phi_n^i(^3P^o)}{E - E_n - k}\langle\phi_n^k(^3P^o)|\, P^k\,|\phi(^3S)\rangle\,. \tag{40}$$

The angular decomposition for the $^3P^o$ reference state is performed by using the identity

$$\begin{aligned} j^i\phi^k &= \frac{1}{3}\,\delta^{ik}\,\vec{j}\cdot\vec{\phi} + \frac{1}{2}\,\epsilon_{ikl}(\vec{j}\times\vec{\phi})_l \\ &\quad + \frac{1}{2}\left(j^i\phi^k + j^k\phi^i - \frac{2}{3}\,\delta^{ik}\,\vec{j}\cdot\vec{\phi}\right). \end{aligned} \tag{41}$$

The three terms in the right-hand side of the above expression give rise to contributions from the ^3S, $^3P^e$, and $^3D^e$

intermediate states, respectively,

$$\begin{aligned} P(k) &= P_{L=0}(k) + P_{L=1}(k) + P_{L=2}(k) \\ &= \frac{1}{3}\langle\Psi_0|\left(\frac{1}{E-H-k}\right)_{^3S}|\Psi_0\rangle \\ &\quad + \frac{1}{2}\langle\Psi_1^i|\left(\frac{1}{E-H-k}\right)_{^3P^e}|\Psi_1^i\rangle \\ &\quad + \frac{1}{4}\langle\Psi_2^{ik}|\left(\frac{1}{E-H-k}\right)_{^3D^e}|\Psi_2^{ik}\rangle, \end{aligned} \tag{42}$$

where $\Psi_0 = \vec{j}\cdot\vec{\phi}$, $\vec{\Psi}_1 = \vec{j}\times\vec{\phi}$, and $\Psi_2^{ik} = j^i\phi^k + j^k\phi^i - \frac{2}{3}\delta^{ik}(\vec{j}\cdot\vec{\phi})$.

A more complicated situation arises in the evaluation of the symmetric part of the E_{L3} contribution for the ^3P reference state [the first term in brackets in Eq. (24), $P_{L3}^{\rm sym}$]. In order to perform the angular decomposition in this case, we use the following identity:

$$\frac{1}{2}\sum_{a=1,2}\left(r_a^i p_a^j + r_a^j p_a^i\right)\phi^k = T^{ijk} + \epsilon^{ikl}\,T^{lj} + \epsilon^{jkl}\,T^{li} + \delta^{ik}\,T^j + \delta^{jk}\,T^i + \delta^{ij}\,T'^k\,, \tag{43}$$

where T^{ijk}, T^{ij}, and T^i are the components of the (symmetric and traceless) irreducible tensors of the first, second, and third rank, respectively,

$$T^{ijk} = \sum_a \left(r_a^i\,p_a^j\,\phi^k\right)^{(3)}, \tag{44}$$

$$T^{ij} = \frac{1}{12}\sum_a\left[\epsilon^{jlm}\left(r_a^i\,p_a^l + r_a^l\,p_a^i\right)\phi^m + \epsilon^{ilm}\left(r_a^j\,p_a^l + r_a^l\,p_a^j\right)\phi^m\right], \tag{45}$$

$$T^i = \frac{1}{20}\sum_a\left[3\left(r_a^i\,p_a^l + r_a^l\,p_a^i\right)\phi^l - 2\,r_a^l\,p_a^l\,\phi^i\right], \tag{46}$$

$$T'^i = \frac{1}{10}\sum_a\left[4r_a^l\,p_a^l\,\phi^i - r_a^i\,p_a^l\,\phi^l - r_a^l\,p_a^i\,\phi^l\right]. \tag{47}$$

Using this identity, we express $P_{L3}^{\rm sym}$ as a sum of the $L=1$, $L=2$, and $L=3$ parts,

$$P_{L3}^{\rm sym}(k) = P_{L3,1}^{\rm sym}(k) + P_{L3,2}^{\rm sym}(k) + P_{L3,3}^{\rm sym}(k), \tag{48}$$

where

$$P_{L3,1}^{\rm sym}(k) = \frac{3\,k^2}{2}\,\frac{4}{3}\,\langle T^i|\left(\frac{1}{E-H-k}\right)_{^3P^o}|T^i\rangle\,, \tag{49}$$

$$P_{L3,2}^{\rm sym}(k) = \frac{3\,k^2}{2}\,\frac{6}{5}\,\langle T^{ij}|\left(\frac{1}{E-H-k}\right)_{^3D^o}|T^{ij}\rangle\,, \tag{50}$$

$$P_{L3,3}^{\rm sym}(k) = \frac{3\,k^2}{2}\,\frac{1}{5}\,\langle T^{ijk}|\left(\frac{1}{E-H-k}\right)_{^3F^o}|T^{ijk}\rangle\,. \tag{51}$$

Wave functions of the different symmetries in Cartesian coordinates required in this work are summarized in Appendix D.

C. Numerical details

Numerical evaluation of the relativistic corrections to the Bethe logarithm was performed according to Eqs. (14), (20), and (27). The general scheme of the computation was similar to the one developed in our previous calculation of the helium fine structure [23] (as described in Sec. V E of that work). Numerical cancelations, however, were much larger in the present work, because of a greater number of asymptotic expansion terms that needed to be separated out.

The low-energy part of the k integral, $k \in (0, K)$ with $K = 10$–100, was evaluated analytically after diagonalizing the matrix representation of the Schrödinger Hamiltonian. In order to perform the high-energy part of the integral, $k \in (K, \infty)$, we calculated the integrand for several hundreds different values of $k \in (5, 10\,000)$, subtracted the contributions of the known asymptotic expansion coefficients, fitted the residual, and calculated the integral analytically. For fitting of the subtracted integrands w_{Li},

$$w_{L1}(k) = k\,P_{L1}(k) - A_1 - \frac{B_1}{\sqrt{k}} - \frac{C_1\ln k}{k} - \frac{D_1}{k}\,, \tag{52}$$

$$w_{L2}(k) = k\,P_{L2}(k) - A_2 - \frac{B_2}{\sqrt{k}} - \frac{C_2\ln k}{k} - \frac{D_2}{k}\,, \tag{53}$$

$$w_{L3}(k) = k\,P_{L3}(k) - G_3k^2 - F_3k - A_3 - \frac{B_3}{\sqrt{k}} - \frac{C_3\ln k}{k} - \frac{D_3}{k}\,, \tag{54}$$

we assumed the following functional forms of their large-k expansion [27]:

$$w_{L1}(k) = \frac{1}{k}\sum_{m=1}^{M}\sum_{n=0}^{m}\frac{c_{m,n}\ln^n k}{k^{m/2}}\,, \tag{55}$$

$$w_{L2,3}(k) = \frac{1}{k}\sum_{m=1}^{M}\frac{d_{m,2}\sqrt{k} + d_{m,1}\ln k + d_{m,0}}{k^m}\,, \tag{56}$$

where $c_{i,j}$ and $d_{i,j}$ are fitting coefficients. In order to ensure the stability of the fitting, high numerical accuracy of the integrand $P_{Li}(k)$ was required, typically 10–12 significant digits.

Such accuracy turned out to be difficult to reach for the perturbed wave-function part of the Breit correction for the 2^3P state. The reason for this is the logarithmic singularity [27] of the perturbed wave function $\delta\phi$,

$$\delta\phi = \frac{1}{(E-H)'}\,H_A^{(4)}\phi\,. \tag{57}$$

In order to ensure good convergence of numerical results for $\delta\phi$, we had to choose the basis for the propagator very carefully. It was constructed as follows. We start by variationally optimizing two symmetric second-order corrections,

$$\delta_1 E = \left\langle H_A^{(4)}\frac{1}{(E-H)'}\,H_A^{(4)}\right\rangle, \tag{58}$$

$$\delta_2 E = \left\langle P^2\frac{1}{(E-H)'}\,P^2\right\rangle. \tag{59}$$

The form of $\delta_2 E$ is suggested by the expression for the leading asymptotic constant A_1, Eq. (A2). In order to account for the logarithmic singularity present in $\delta_1 E$, we exploit

TABLE I. Numerical results for the relativistic corrections to the Bethe logarithm and asymptotic expansion constants for the 2^3S and 2^3P (centroid) states of helium, in atomic units.

Term	2^3S	2^3P
D	33.184 142 629	31.638 617 831 (1)
A_1	−33.989 031 782 (2)	−31.977 565 646
D_1	−132.158 242 69 (5)	−127.498 493 92 (10)
E_{L1}	−45.1291 (35)	−41.7175 (40)
A_2	42.692 780 038	40.253 149 916
D_2	−53.768 709 997 (3)	−50.260 445 50 (9)
E_{L2}	335.8675 (36)	319.1601 (36)
G_3	0.032 569 625	−0.065 018 180
F_3	2.121 589 807	2.079 835 929
A_3	−49.768 158 799	−47.453 391 8 (3)
D_3	1 175.043 968 722 (4)	1 121.176 717 34 (10)
E_{L3}	−1 095.043 9 (3)	−1 045.214 (6)

the flexibility of our exponential basis functions (38) and emulate the singularity by allowing the nonlinear parameters to be very large. In order to effectively span large regions of nonlinear parameters, we used a nonuniform distribution of nonlinear parameters α_i, β_i, and γ_i introduced in Ref. [33], typically,

$$\alpha_i = A_1 + \left(1/t_i^3 - 1\right) A_2\,, \tag{60}$$

where the variable t_i has a uniform quasirandom distribution over the interval (0,1), and A_1 and A_2 are the variational optimization parameters. Finally, we merge the optimized basis sets for $\delta_1 E$ and $\delta_2 E$ and use the result for calculating the perturbed wave function $\delta\phi$. Nonetheless, a large number of basis functions ($N = 3000$–5000) were required in order to reach the desired accuracy.

V. RESULTS

Our numerical results for the asymptotic expansion coefficients and the relativistic corrections to the Bethe logarithm are presented in Table I. For the 2^3S–2^3P transition energy in helium, the total relativistic correction to the Bethe logarithm, $E_L = E_{L1} + E_{L2} + E_{L3}$, amounts to $E_L = -4.9743\,(13)$ MHz. This can be compared with the estimate of Refs. [34] and [25], obtained from the hydrogenic results by rescaling the electron density at the origin. For the 2^3S–2^3P transition energy, this approximation yields

$$E_L(\text{appr}) = \alpha^7\, m\, Z^3 \left[\mathcal{L}(2s) - \tfrac{1}{3}\mathcal{L}(2p_{1/2}) - \tfrac{2}{3}\mathcal{L}(2p_{3/2})\right] \times [\langle\delta^3(r_1) + \delta^3(r_2)\rangle_{2^3S} - \langle\delta^3(r_1) + \delta^3(r_2)\rangle_{2^3P}], \tag{61}$$

with $\mathcal{L}(2s) = -28.350\,965$, $\mathcal{L}(2p_{1/2}) = -0.795\,650$, and $\mathcal{L}(2p_{3/2}) = -0.584\,517$ [35,36]. The corresponding numerical value is $E_L(\text{appr}) = -3.7\,(0.9)$ MHz, where we assumed a 25% uncertainty, like in Ref. [25].

A complete treatment of the $\alpha^7 m$ correction requires calculations of the two remaining terms in Eq. (1), which will be addressed in our future investigations. The numerical contribution of these terms is expected to be comparable to that of E_L. In particular, for the 2^3S–2^3P transition energy in helium, the hydrogenic approximation for the remaining contribution yields $-4.3\,(1.1)$ MHz.

In summary, in this work we report calculations of the relativistic corrections to the Bethe logarithm for the $2\,^3S$ and $2\,^3P$ states of helium. This is the first step on the path to calculating the complete QED contribution of order $\alpha^7 m$ to the triplet states of helium. Being the most numerically demanding part, the calculation of the relativistic corrections to the Bethe logarithm indicates the feasibility of the whole $\alpha^7\, m$ project.

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APPENDIX A: ASYMPTOTIC COEFFICIENTS OF P_{L1}

Here we derive the coefficients A_1, B_1, C_1, and D_1 of the large-k expansion of P_{L1} given by Eq. (12). There are contributions coming from both the low-energy and the high-energy regions of the virtual photon momenta. Individually, the low- and the high-energy parts may contain divergences, which are regularized by working in d dimensions and are canceled when both parts are added together.

1. Low-energy part

The low-energy part can be derived by performing a direct large-k expansion of the expression

$$\delta\left\langle P^i \frac{k}{E - H - k} P^i\right\rangle, \tag{A1}$$

where δ denotes the first-order perturbation of the matrix element by Breit Hamiltonian $H^{(4)}$. The coefficient A_1 from Eq. (12) comes from the perturbation of the reference-state wave function,

$$\begin{aligned} A_1 &= -2\left\langle H^{(4)} \frac{1}{(E - H)'} P^2\right\rangle \\ &= -2\left\langle H_A^{(4)} \frac{1}{(E - H)'}\, P^2\right\rangle + 2\langle[Q - \langle Q\rangle]\, P^2\rangle\,. \end{aligned} \tag{A2}$$

The low-energy part of the coefficient D_1 is

$$\begin{aligned} D_1^L &= \delta\,\langle P^i(H - E)P^i\rangle \\ &= \left\langle H^{(4)} \frac{1}{(E - H)'} [P^i, [V, P^i]]\right\rangle + \frac{1}{2}\langle[P^i, [H^{(4)}, P^i]]\rangle \\ &= D_{1a}^L + D_{1b}^L. \end{aligned} \tag{A3}$$

The second-order term D_{1a}^L is singular. We employ the regularized form of the Breit Hamiltonian $H_A^{(4)}$, Eq. (28), in order to move singularities into first-order terms and use the dimensional regularization in order to handle the remaining divergences. The result is

$$D^L_{1a} = -2\,Z\left\langle H^{(4)}_A \frac{1}{(E-H)'}\left(\frac{\vec{r}_1}{r_1^3}\cdot\vec{\nabla}_1 + \frac{\vec{r}_2}{r_2^3}\cdot\vec{\nabla}_2\right)\right\rangle + E^{(4)}\left(\left\langle\frac{2}{r}\right\rangle - 4E\right)$$
$$+\left\langle\left[\frac{Z}{r_1}\right]_\epsilon\left(E + \left[\frac{Z}{r_1}+\frac{Z}{r_2}\right]_\epsilon - \frac{1}{r}\right)^2 + \frac{1}{4}\left[\frac{Z^2}{r_1^4}\right]_\epsilon - \frac{1}{2}\,p_1^2\frac{Z}{r_1}\,p_2^2\right.$$
$$\left.+\left(2E - \frac{2}{r_2} + \left\langle\frac{1}{r}\right\rangle\right)\pi Z\delta^{(3)}(r_1) + p_1^i\,\frac{Z}{r_1}\left(\frac{\delta^{ij}}{r} + \frac{r^i r^j}{r^3}\right)p_2^j + (1\leftrightarrow 2)\right\rangle. \tag{A4}$$

Here, $[Z/r_1]_\epsilon$ is the d-dimensional form of the Coulomb potential (for details see [20]). The terms $[(Z/r_1)^3]_\epsilon$ and $[Z^2/r_1^4]_\epsilon$ contain singularities which will be canceled when combined with corresponding terms coming from the high-energy part. Term D^L_{1b} is evaluated as

$$D^L_{1b} = \left\langle -\left(E + \frac{Z-1}{r_2} - \frac{p_2^2}{2}\right)\pi Z\,\delta^{(3)}(r_1) + \frac{Z}{2}\,\vec{p}_1\,\pi\,\delta^{(3)}(r_1)\,\vec{p}_1 + (1\leftrightarrow 2)\right\rangle. \tag{A5}$$

2. High-energy part

Coefficient B_1 can be obtained from the forward-scattering two-photon exchange diagram perturbed by the Breit Hamiltonian. The result is

$$B_1 = \sqrt{2}\,Z^2\langle\pi\left[\delta^{(3)}(r_1) + \delta^{(3)}(r_2)\right]\rangle. \tag{A6}$$

The Breit correction to the forward-scattering three-photon exchange diagram contains both the coefficient C_1 and the high-energy part of the coefficient D_1,

$$C_1 = Z^3\,\langle\pi\left[\delta^{(3)}(r_1) + \delta^{(3)}(r_2)\right]\rangle, \tag{A7}$$

and

$$D^H_1 = Z^3\,\langle\pi\left[\delta^{(d)}(r_1) + \delta^{(d)}(r_2)\right]\rangle\left(-8 - \frac{1}{2\epsilon} + 9\ln 2\right). \tag{A8}$$

Finally, the coefficient D_1 is the sum of the low-energy part D^L_1 and the high-energy part D^H_1. Making use of the identity

$$\left[\frac{Z^2}{r_1^4}\right]_\epsilon = -2\left[\frac{Z^3}{r_1^3}\right]_\epsilon + \vec{p}_1\,\frac{Z^2}{r_1^2}\,\vec{p}_1 + p_2^2\frac{Z^2}{r_1^2} - 2\left(E + \frac{Z}{r_2} - \frac{1}{r}\right)\frac{Z^2}{r_1^2}, \tag{A9}$$

we write the result as

$$D_1 = -2\,Z\left\langle H^{(4)}_A\frac{1}{(E-H)'}\left(\frac{\vec{r}_1}{r_1^3}\cdot\vec{\nabla}_1 + \frac{\vec{r}_2}{r_2^3}\cdot\vec{\nabla}_2\right)\right\rangle + E^{(4)}\left(\left\langle\frac{2}{r}\right\rangle - 4E\right)$$
$$+\left\langle\frac{Z}{2}\,\vec{p}_1\,\pi\,\delta^{(3)}(r_1)\,\vec{p}_1 + \frac{Z}{r_1}(E-V)^2 - \frac{X_1}{2} - \frac{1}{2}\,p_1^2\,\frac{Z}{r_1}\,p_2^2 + p_1^i\,\frac{Z}{r_1}\left(\frac{\delta^{ij}}{r} + \frac{r^i r^j}{r^3}\right)p_2^j\right.$$
$$\left.+\left[E - \frac{Z+1}{r_2} + \frac{p_2^2}{2} + \left\langle\frac{1}{r}\right\rangle + Z^2(-7 + 9\ln 2)\right]\pi Z\,\delta^{(3)}(r_1) + (1\leftrightarrow 2)\right\rangle, \tag{A10}$$

where

$$\left\langle\left[\frac{Z^3}{r_1^3}\right]_\epsilon\right\rangle = \left\langle\frac{1}{r_1^3}\right\rangle + Z^3\langle\pi\,\delta^d(r_1)\rangle\left(\frac{1}{\epsilon} + 2\right), \tag{A11}$$

$$\left\langle\frac{1}{r_1^3}\right\rangle = \lim_{a\to 0}\left\langle\frac{\Theta(r_1 - a)}{r_1^3} + 4\,\pi\,\delta^3(r_1)\,(\gamma + \ln a)\right\rangle, \tag{A12}$$

$$X_1 = \frac{Z^2}{r_1^2}\left(E - V - \frac{p_2^2}{2}\right) - \frac{1}{2}\,\vec{p}_1\,\frac{Z^2}{r_1^2}\,\vec{p}_1\,. \tag{A13}$$

APPENDIX B: ASYMPTOTIC COEFFICIENTS OF P_{L2}

Here we derive the coefficients A_2, B_2, C_2, and D_2 of the large-k expansion of P_{L2} given by Eq. (18).

1. Low-energy part

First we examine contributions coming from the region of low virtual photon momenta. The coefficient A_2 is the leading-order term of the direct large-k expansion of Eq. (17), with the result

$$A_2 = \frac{1}{2}\left\langle 4\,(E-V)^2 - 2\,p_1^2\,p_2^2 + 2\,(E-V)\,\vec{p}_1\cdot\vec{p}_2 + \left(\frac{\delta^{ij}}{r} + \frac{r^i r^j}{r^3}\right)\left(p_1^i + p_2^i\right)\left(p_1^j + p_2^j\right)\right\rangle. \tag{B1}$$

The low-energy part of the coefficient D_2 is

$$D_2^L = \langle\phi|\delta j^i\,(H-E)\,j^i|\phi\rangle = \left\langle\frac{1}{2}\left[\delta j^i, \left[-\left[\frac{Z}{r_1}+\frac{Z}{r_2}\right]_\epsilon, j^i\right]\right]\right\rangle = D_{2a}^L + D_{2b}^L. \tag{B2}$$

Individual terms are

$$D_{2a}^L = \left\langle\frac{1}{4}\left[\left(p_1^i\,p_1^2 + p_2^i\,p_2^2\right), \left[\left[\frac{Z}{r_1}+\frac{Z}{r_2}\right]_\epsilon, j^i\right]\right]\right\rangle = \left\langle -\left[2\left(E + \frac{Z-1}{r_2}\right) - p_2^2\right]\pi\,Z\,\delta^{(3)}(r_1)\right.$$
$$\left. - \frac{1}{2}\,\vec{p}_1\frac{Z^2}{r_1^2}\vec{p}_1 + \left[\frac{Z}{r_1}\right]_\epsilon^2\left(E + \left[\frac{Z}{r_1}+\frac{Z}{r_2}\right]_\epsilon - \frac{1}{r}\right) - \frac{1}{2}\,p_2^2\frac{Z^2}{r_1^2} + \frac{1}{2}\frac{Z\vec{r}_1}{r_1^3}\cdot\frac{\vec{r}}{r^3} + (1\leftrightarrow 2)\right\rangle, \tag{B3}$$

and

$$D_{2b}^L = \left\langle\frac{1}{4}\left[\left(\frac{\delta^{ij}}{r}+\frac{r^i r^j}{r^3}\right)\left(p_1^j + p_2^j\right), \left[\frac{Z}{r_1}+\frac{Z}{r_2}, j^i\right]\right]\right\rangle$$
$$= \left\langle\frac{Z}{4}\left(\frac{\delta^{ij}}{r}+\frac{r^i r^j}{r^3}\right)\left(\frac{3r_1^i r_1^j - \delta^{ij}\,r_1^2}{r_1^5}\right) - \frac{4\pi}{3}\frac{Z}{r_2}\,\delta^{(3)}(r_1) + (1\leftrightarrow 2)\right\rangle. \tag{B4}$$

2. High-energy part

Now we turn to contributions induced by high momenta of virtual photons. The coefficient B_2 comes from the forward-scattering two-photon exchange perturbed by δj^i and can be evaluated to yield

$$B_2 = -\,Z^2\,\sqrt{2}\,\langle 4\pi\,[\delta^3(r_1) + \delta^3(r_2)]\rangle. \tag{B5}$$

Similarly to the case of P_{L1}, the coefficient C_2 and the high-energy part of D_2 are obtained from the forward-scattering three-photon exchange with additional δj^i and j^i vertices,

$$\phi^2(0)\int\frac{d^d q_1^2}{(2\pi)^d}\frac{d^d q_2^2}{(2\pi)^d}\left(\frac{-4\pi Z}{q_1^2}\right)\left(\frac{-4\pi Z}{q_2^2}\right)\left(\frac{-4\pi Z}{(\vec{q}_1-\vec{q}_2)^2}\right)\left(\frac{2}{q_1^2}\right)\left(\frac{2}{q_2^2}\right)\left[\frac{-\frac{1}{2}q_1^2\,(\vec{q}_1\cdot\vec{q}_2)}{\left(\frac{q_1^2}{2}+k\right)\left(\frac{q_2^2}{2}+k\right)} - \frac{q_2^2}{\left(\frac{q_2^2}{2}+k\right)} - \frac{q_1^2}{\left(\frac{q_1^2}{2}+k\right)}\right]. \tag{B6}$$

From this expression, we derive the following results:

$$C_2 = \frac{Z^3}{2}\,\langle 4\pi\,[\delta^3(r_1) + \delta^3(r_2)]\rangle \tag{B7}$$

and

$$D_2^H = \left\langle Z^3\,\pi\left[\delta^{(d)}(r_1) + \delta^{(d)}(r_2)\right]\right\rangle\left(8 - \frac{1}{\epsilon} - 6\ln 2\right). \tag{B8}$$

The total coefficient D_2 is then the sum of the corresponding low-energy and high-energy parts,

$$D_2 = \left\langle\frac{Z}{4}\left(\frac{\delta^{ij}}{r}+\frac{r^i r^j}{r^3}\right)\left(\frac{3r_1^i r_1^j - \delta^{ij}\,r_1^2}{r_1^5}\right) - \left(\frac{E}{2} + \frac{3Z-1}{6r_2} - Z^2\frac{5-3\ln 2}{2} - \frac{p_2^2}{4}\right)4\pi\,Z\,\delta^{(3)}(r_1)\right.$$
$$\left. + X_1 + \frac{Z}{2}\frac{\vec{r}_1}{r_1^3}\cdot\frac{\vec{r}}{r^3} + (1\leftrightarrow 2)\right\rangle. \tag{B9}$$

APPENDIX C: ASYMPTOTIC COEFFICIENTS OF P_{L3}

We now turn to the derivation of the coefficients G_3, F_3, A_3, B_3, C_3, and D_3 of the large-k expansion of P_{L3} given by Eq. (25), which is the most complicated part.

1. Low-energy part

In order to derive contributions coming from the low photon momenta, we first make a large-k expansion of the propagator $1/(E-H-k)$ in Eq. (22). In the obtained expression, we then make a small-k expansion and keep the k^2 contribution.

Using the angular average identity in d dimensions (with $\hat{k}=\vec{k}/k$)

$$\int \frac{d\Omega_k}{4\pi}\,\hat{k}^m\hat{k}^n(\delta^{ij}-\hat{k}^i\hat{k}^j)=\frac{1}{d\,(d+2)}[(d+1)\,\delta^{ij}\delta^{mn}-\delta^{im}\delta^{jn}-\delta^{in}\delta^{jm}], \tag{C1}$$

we get

$$G_3=-\frac{3}{2}\sum_{a,b=1,2}\int\frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\,\delta'_{k^2}\big\langle p_a^i\,e^{-i\vec{k}\cdot(\vec{r}_a-\vec{r}_b)}\,p_b^j\big\rangle=\frac{1}{5}\,\big\langle p_1^i(2\delta^{ij}r^2-r^ir^j)p_2^j\big\rangle, \tag{C2}$$

where the symbol δ'_{k^2} stands for performing a small-k expansion and taking the coefficient at the k^2 term.

Analogously, the next coefficient F_3 is obtained as

$$F_3=\frac{3}{2}\sum_{a,b=1,2}\int\frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\big\langle p_a^i\,e^{-i\vec{k}\cdot\vec{r}_a}\,(H-E)\,p_b^j\,e^{i\vec{k}\cdot\vec{r}_b}\big\rangle=\Big\langle E-V-\frac{1}{5r}\Big\rangle. \tag{C3}$$

Furthermore,

$$\begin{aligned}A_3=&-\frac{3}{2}\sum_{a,b=1,2}\int\frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\,\big\langle p_a^i\,e^{-i\vec{k}\cdot\vec{r}_a}\,(H-E)^2\,p_b^j\,e^{i\vec{k}\cdot\vec{r}_b}\big\rangle\\=&\;\frac{1}{10}\Big\langle Z^2\frac{4\,(\vec{r}_1\cdot\vec{r}_2)\,r^2-2\,(\vec{r}_1\cdot\vec{r})\,(\vec{r}_2\cdot\vec{r})}{r_1^3r_2^3}-\frac{2}{r^2}-\frac{4}{5}\,(E-V)^2-6\,(\vec{p}_1\cdot\vec{p}_2)^2+6\,p_1^2\,p_2^2\\&+\Big[p_2^j\Big(\frac{Zr_1^i}{r_1^3}-\frac{r^i}{r^3}\Big)(3\,\delta^{ik}r^j+3\,\delta^{ij}r^k-2\,\delta^{jk}r^i)\,p_2^k+2\frac{Z\vec{r}_1}{r_1^3}\cdot\frac{\vec{r}}{r}-2\pi\,Z\,\delta^3(r_1)+(1\leftrightarrow 2)\Big]\Big\rangle.\end{aligned} \tag{C4}$$

The low-energy part of the coefficient D_3 is the most complicated term and thus will be discussed in some detail. The starting expression is

$$D_3^L=\frac{3}{2}\sum_{a,b=1,2}\int\frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\,\big\langle p_a^i\,e^{-i\vec{k}\cdot\vec{r}_a}\,(H-E)^3\,p_b^j\,e^{i\vec{k}\cdot\vec{r}_b}\big\rangle. \tag{C5}$$

It is convenient to split the above expression into two parts, with $a=b$ and $a\neq b$. The first part can be evaluated with help of the identity

$$e^{-i\vec{k}\cdot\vec{r}}\,f(p)\,e^{i\vec{k}\cdot\vec{r}}=f(p+k)\,. \tag{C6}$$

We obtain

$$D_{3a}^L=\frac{3}{2}\sum_{a=1,2}\int\frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\,\big\langle p_a^i\,\big(H-E+\vec{p}_a\cdot\vec{k}+k^2/2\big)^3\,p_a^j\big\rangle. \tag{C7}$$

After straightforward but tedious manipulations that involve expanding the matrix element in small k and retaining the coefficient in front of k^2 and using identities

$$\begin{aligned}&\Big[p_1^2,\Big[p_1^2,\frac{Z}{r_1}\Big]\Big]=4\Big[\frac{Z^2}{r_1^4}\Big]_\epsilon-4\frac{Z\vec{r}_1\cdot\vec{r}}{r_1^3r^3},\quad\Big[p_1^2,\Big[p_1^2,\frac{1}{r}\Big]\Big]=2\frac{Z\vec{r}_1\cdot\vec{r}}{r_1^3r^3}-\frac{2}{r^4}-P^iP^j\frac{3r^ir^j-\delta^{ij}r^2}{r^5},\\&p_1^i\,[\,p_1^i,[V,p_1^j]]\,p_1^j=\Big[\frac{Z^2}{r_1^4}\Big]_\epsilon-\frac{3}{2}\,\frac{Z\vec{r}_1\cdot\vec{r}}{r_1^3r^3}+\frac{1}{2\,r^4}+\frac{1}{4}\,P^iP^j\,\frac{3r^ir^j-\delta^{ij}r^2}{r^5}+\frac{1}{2}\,p_1^i\,[\,p_1^j,[V,p_1^j]]\,p_1^i\\&\qquad-\Big(E+\frac{Z}{r_2}-\frac{1}{r}-\frac{p_2^2}{2}\Big)4\pi\,Z\,\delta^{(3)}(r_1)\,,\end{aligned} \tag{C8}$$

as well as Eq. (A9), we arrive at

$$\begin{aligned}D_{3a}^L=&\;\frac{3}{2}\Big\langle\frac{7}{6}\,\frac{1}{r^4}+\frac{1}{12}\,P^iP^j\,\frac{3r^ir^j-\delta^{ij}r^2}{r^5}+\frac{1}{10}\,\vec{p}_1\,4\pi\,Z\,\delta^{(3)}(r_1)\,\vec{p}_1-\frac{5}{2}\,\frac{Z\vec{r}_1\cdot\vec{r}}{r_1^3r^3}+\frac{4}{3}\,\vec{p}_1\,\frac{Z^2}{r_1^2}\,\vec{p}_1-\frac{8}{3}\,\frac{Z^2}{r_1^2}(E-V)\\&+\frac{4}{3}\,p_2^2\frac{Z^2}{r_1^2}+\Big[\frac{28}{15}\Big(E+\frac{Z}{r_2}-\frac{1}{r}-\frac{p_2^2}{2}\Big)+Z^2\Big(-\frac{8}{3\,\epsilon}-\frac{206}{45}\Big)\Big]\pi\,Z\,\delta^{(d)}(r_1)-\frac{1}{10}\,\vec{p}_1\,4\pi\,\delta^{(3)}(r)\,\vec{p}_1+(1\leftrightarrow 2)\Big\rangle.\end{aligned} \tag{C9}$$

The second term in Eq. (C5) with $a \neq b$ is evaluated as

$$\begin{aligned} D^L_{3b} &= \frac{3}{2}\int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\,\delta'_{k^2}\big\langle p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1}\,(H-E)^3\,p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\big\rangle + (1\leftrightarrow 2)\\ &= \frac{3}{4}\int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\,\delta'_{k^2}\big\langle\big[\big[p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1},\,H-E\big],\big[H-E,\big[H-E,\;p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\big]\big]\big]\big\rangle + (1\leftrightarrow 2) = \frac{3}{4}\sum_{m=1..8} T_m\,. \end{aligned} \tag{C10}$$

The individual terms T_i are calculated as follows:

$$T_1 = \int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\big\langle\big[\big[p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1},\,V\big],\big[V,\big[V,\;p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\big]\big]\big]\big\rangle + (1\leftrightarrow 2) = 0\,, \tag{C11}$$

$$\begin{aligned} T_2 &= \int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\bigg\langle\bigg[\big[p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1},\,V\big],\bigg[V,\bigg[\frac{p_2^2}{2},\;p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\bigg]\bigg]\bigg]\bigg\rangle + (1\leftrightarrow 2)\\ &= \frac{2}{15}\bigg[\bigg(\frac{Z\vec{r}_1}{r_1^3}-\frac{Z\vec{r}_2}{r_2^3}\bigg)\cdot\frac{\vec{r}}{r^3}-\frac{2}{r^4}\bigg], \end{aligned} \tag{C12}$$

$$T_3 = \int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\bigg\langle\bigg[\bigg[p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1},\,\frac{p_1^2}{2}\bigg],\big[V,\big[V,\;p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\big]\big]\bigg]\bigg\rangle + (1\leftrightarrow 2) = 0\,, \tag{C13}$$

$$\begin{aligned} T_4 &= \int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\bigg\langle\bigg[\big[p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1},\,V\big],\bigg[\frac{p_1^2}{2}+\frac{p_2^2}{2},\big[V,\;p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\big]\bigg]\bigg]\bigg\rangle + (1\leftrightarrow 2)\\ &= \frac{4}{15}\bigg(\frac{2}{r^4}+\bigg[2Z\bigg(\frac{\delta^{ij}}{r_1^3}-3\frac{r_1^ir_1^j}{r_1^5}\bigg)\frac{r^ir^j}{r^3}+3\,\frac{Z\vec{r}_1}{r_1^3}\cdot\frac{\vec{r}}{r^3}-\frac{1}{3r_2}4\pi Z\,\delta^{(3)}(r_1)+(1\leftrightarrow 2)\bigg]\bigg), \end{aligned} \tag{C14}$$

$$\begin{aligned} T_5 &= \int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\bigg\langle\bigg[\bigg[p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1},\,\frac{p_1^2}{2}\bigg],\bigg[V,\bigg[\frac{p_2^2}{2},\;p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\bigg]\bigg]\bigg]\bigg\rangle + (1\leftrightarrow 2)\\ &= -\frac{16}{45}\,\vec{p}\,4\pi\,\delta^{(3)}(r)\,\vec{p}+\frac{P^iP^j}{15}\bigg(\frac{\delta^{ij}}{r^3}-3\frac{r^ir^j}{r^5}\bigg)-\frac{4}{15}\,p^i\bigg(\frac{\delta^{ij}}{r^3}-3\frac{r^ir^j}{r^5}\bigg)\,p^j\,, \end{aligned} \tag{C15}$$

where $p^i = \frac{1}{2}(p_1^i - p_2^i)$,

$$\begin{aligned} T_6 &= \int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\bigg\langle\bigg[\big[p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1},\,V\big],\bigg[\frac{p_2^2}{2},\bigg[\frac{p_2^2}{2},\;p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\bigg]\bigg]\bigg]\bigg\rangle + (1\leftrightarrow 2)\\ &= -\frac{4}{45}\,\vec{p}\,4\pi\,\delta^{(3)}(r)\,\vec{p}+\frac{2P^iP^j}{15}\bigg(\frac{\delta^{ij}}{r^3}-3\frac{r^ir^j}{r^5}\bigg)+\frac{8}{15}\,p^i\bigg(\frac{\delta^{ij}}{r^3}-3\frac{r^ir^j}{r^5}\bigg)p^j\,, \end{aligned} \tag{C16}$$

$$\begin{aligned} T_7 &= \int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\bigg\langle\bigg[\bigg[p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1},\,\frac{p_1^2}{2}\bigg],\bigg[\frac{p_1^2+p_2^2}{2},\big[V,\;p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\big]\bigg]\bigg]\bigg\rangle + (1\leftrightarrow 2)\\ &= -\frac{8}{45}\,\vec{p}\,4\pi\,\delta^{(3)}(r)\,\vec{p}+\frac{2P^iP^j}{15}\bigg(\frac{\delta^{ij}}{r^3}-3\frac{r^ir^j}{r^5}\bigg)+\frac{16}{15}\,p^i\bigg(\frac{\delta^{ij}}{r^3}-3\frac{r^ir^j}{r^5}\bigg)p^j\,, \end{aligned} \tag{C17}$$

and

$$T_8 = \int \frac{d\Omega_k}{4\pi}\,(\delta^{ij}-\hat{k}^i\hat{k}^j)\delta'_{k^2}\bigg\langle\bigg[\bigg[p_1^i\,e^{-i\vec{k}\cdot\vec{r}_1},\,\frac{p_1^2}{2}\bigg],\bigg[\frac{p_1^2+p_2^2}{2},\bigg[\frac{p_2^2}{2},\;p_2^j\,e^{i\vec{k}\cdot\vec{r}_2}\bigg]\bigg]\bigg]\bigg\rangle + (1\leftrightarrow 2) = 0\,. \tag{C18}$$

Substituting the terms T_m into Eq. (C10), we obtain the result for the low-energy part of the coefficient D_3,

$$\begin{aligned} D_3^L = 3\bigg\langle &\frac{1}{3}\,p^i\bigg(\frac{\delta^{ij}}{r^3}-3\frac{r^ir^j}{r^5}\bigg)p^j-\frac{23}{90}\,\vec{p}\,4\pi\,\delta^{(3)}(r)\,\vec{p}+\frac{37}{30}\,\frac{1}{r^4}+\frac{1}{2}\bigg\{-\frac{8}{3}\,\frac{Z^2}{r_1^2}(E-V)+\frac{4}{3}\,\vec{p}_1\,\frac{Z^2}{r_1^2}\,\vec{p}_1\\ &+\frac{1}{10}\,\vec{p}_1\,4\pi Z\,\delta^{(3)}(r_1)\,\vec{p}_1-\frac{61}{30}\,\frac{Z\vec{r}_1\cdot\vec{r}}{r_1^3r^3}+\frac{4}{3}\,p_2^2\frac{Z^2}{r_1^2}+\frac{4}{15}Z\bigg(\frac{\delta^{ij}}{r_1^3}-3\frac{r_1^ir_1^j}{r_1^5}\bigg)\frac{r^ir^j}{r^3}\\ &+\bigg[\frac{7}{15}\bigg(E+\frac{Z}{r_2}-\frac{p_2^2}{2}\bigg)-\frac{23}{45}\frac{1}{r_2}-Z^2\bigg(\frac{2}{3\,\epsilon}+\frac{103}{90}\bigg)\bigg]4\pi Z\,\delta^{(d)}(r_1)+(1\leftrightarrow 2)\bigg\}\bigg\rangle. \end{aligned} \tag{C19}$$

2. High-energy part

The coefficient B_3 comes from the corresponding forward-scattering two-photon exchange diagram,

$$B_3 = \frac{3k\sqrt{k}}{2}\int\frac{d\Omega_k}{4\pi}\,\delta_{k^2}\,\phi^2(0)\int\frac{d^3p}{(2\pi)^3}\left[\frac{-4\pi\,Z\alpha}{p^2}\right]^2\left(\frac{2}{p^2}\right)^2 p^i\,\frac{(-2)}{(p+k)^2+2\omega}\,p^j\,(\delta^{ij}-\hat{k}^i\hat{k}^j)$$
$$= 2\sqrt{2}\,Z^2\,\langle 4\pi[\delta^3(r_1)+\delta^3(r_2)]\rangle\,. \tag{C20}$$

Similarly, the analogous forward-scattering three-photon exchange diagram gives rise to the coefficient C_3,

$$C_3 = -2\,Z^3\,\langle 4\pi[\delta^3(r_1)+\delta^3(r_2)]\rangle\,, \tag{C21}$$

and the high-energy part of the coefficient D_3,

$$D_3^H = Z^3\,\big\langle 4\pi\big[\delta^{(d)}(r_1)+\delta^{(d)}(r_2)\big]\big\rangle\left(-\frac{73}{45}+\frac{2}{3\,\epsilon}+\frac{4}{3}\ln 2\right). \tag{C22}$$

The total coefficient D_3 is obtained as a sum of the low-energy part D_3^L and the high-energy part D_3^H. Using the identity,

$$p^i\left(\frac{\delta^{ij}}{r^3}-3\frac{r^ir^j}{r^5}\right)p^j = \frac{2\pi}{3}\,\vec{p}\,\delta^{(3)}(r)\,\vec{p}-\frac{1}{2\,r^4}+\frac{1}{4}\left(\frac{Z\vec{r}_1}{r_1^3}-\frac{Z\vec{r}_2}{r_2^3}\right)\cdot\frac{\vec{r}}{r^3}\,, \tag{C23}$$

we express the final result for D_3 as

$$D_3 = \frac{1}{5}\bigg\langle -3\,\vec{p}\;4\pi\,\delta^{(3)}(r)\,\vec{p}+\frac{16}{r^4}+\bigg[2\,Z\left(\frac{\delta^{ij}}{r_1^3}-3\frac{r_1^ir_1^j}{r_1^5}\right)\frac{r^ir^j}{r^3}-20\,X_1+\frac{3}{4}\,\vec{p}_1\,4\pi\,Z\,\delta^{(3)}(r_1)\,\vec{p}_1-14\,\frac{Z\vec{r}_1}{r_1^3}\cdot\frac{\vec{r}}{r^3}$$
$$+\left(\frac{7E}{2}+\frac{7Z}{2}\frac{1}{r_2}-\frac{23}{6}\frac{1}{r_2}-\frac{7}{4}\,p_2^2-\frac{83Z^2}{4}+10\,Z^2\ln 2\right)4\pi\,Z\,\delta^{(3)}(r_1)+(1\leftrightarrow 2)\bigg]\bigg\rangle. \tag{C24}$$

The operator $\vec{p}_1\,4\pi\,Z\,\delta^{(3)}(r_1)\,\vec{p}_1$ requires some clarifications, because its expectation value is conditionally converging. It should be calculated with the implicit projection into the $L=0$ state between $\vec{p}_1$ operators, and this requirement comes from the dimensional regularization.

APPENDIX D: WAVE FUNCTIONS IN CARTESIAN COORDINATES

Following Schwartz [28], we use the following representations of the wave functions, with $F\equiv F(r_1,r_2,r)$, $G\equiv G(r_1,r_2,r)$, and the upper sign corresponding to the singlet function and the lower sign to the triplet function:

$$\phi(^{1,3}S^e) = F\pm(1\leftrightarrow 2)\,, \tag{D1}$$

$$\vec{\phi}(^{1,3}P^o) = \vec{r}_1\,F\pm(1\leftrightarrow 2)\,, \tag{D2}$$

$$\vec{\phi}(^{1,3}P^e) = \vec{r}_1\times\vec{r}_2\,F\pm(1\leftrightarrow 2)\,, \tag{D3}$$

$$\phi^{ij}(^{1,3}D^o) = \left(\epsilon^{iab}r_1^ar_2^br_1^j+\epsilon^{jab}r_1^ar_2^br_1^i\right)F\pm(1\leftrightarrow 2)\,, \tag{D4}$$

$$\phi^{ij}(^{1,3}D^e) = \left(r_1^ir_1^j-\tfrac{1}{3}\,\delta^{ij}r_1^2\right)F+\tfrac{1}{2}\left(r_1^ir_2^j+r_2^ir_1^j-\tfrac{2}{3}\,\delta^{ij}\,\vec{r}_1\cdot\vec{r}_2\right)G\pm(1\leftrightarrow 2)\,, \tag{D5}$$

and

$$\phi^{ijk}(^{1,3}F^o) = \left[r_1^ir_1^jr_1^k-\tfrac{1}{5}\,r_1^2\left(\delta^{ij}r_1^k+\delta^{ik}r_1^j+\delta^{jk}r_1^i\right)\right]F+\tfrac{1}{3}\left[r_1^ir_1^jr_2^k+r_1^ir_2^jr_1^k+r_2^ir_1^jr_1^k-\tfrac{1}{5}\,\delta^{ij}\left(r_1^2r_2^k+2\,\vec{r}_1\cdot\vec{r}_2r_1^k\right)\right.$$
$$\left.-\tfrac{1}{5}\,\delta^{ik}\left(r_1^2r_2^j+2\,\vec{r}_1\cdot\vec{r}_2r_1^j\right)-\tfrac{1}{5}\,\delta^{jk}\left(r_1^2r_2^i+2\,\vec{r}_1\cdot\vec{r}_2r_1^i\right)\right]G\pm(1\leftrightarrow 2)\,. \tag{D6}$$

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