Breit-Pauli and Direct Perturbation Theory Calculations of Relativistic Helium Polarizability

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Large Gaussian-type geminal wave function expansions and direct perturbation theory (DPT) of relativistic effects have been applied to calculate the relativistic contribution to the static dipole polarizability of the helium atom. It has been demonstrated that DPT is superior for this purpose to traditional Breit-Pauli calculations. The resulting value of the molar polarizability of ^{4}He is 0.517254(1) cm^3 mol^{-1}, including a literature estimate of QED effects. As a by-product, a very accurate value of the nonrelativistic helium second hyperpolarizability, $\gamma = 43.104227(1)$ atomic units (without the mass-polarization correction), has been obtained.

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For light atomic and molecular systems the relativistic effects are usually included by calculating an average value of the Breit-Pauli operator [1] with a nonrelativistic wave function. An alternative approach is the direct perturbation theory (DPT) formulated by Rutkowski [2] and Kutzelnigg [3] and applied recently to simple atomic [4,5] and molecular systems [4,6–8]. In this paper we apply the two approaches in a calculation of the relativistic contribution to a second-order property of crucial experimental importance, the polarizability of helium.

It is well known [9] that the pressure of a gas can be expressed in terms of its temperature, dielectric constant, and molar polarizability $A_\varepsilon = \frac{4}{3}\pi N_A \alpha$, where $N_A = 6.022\,142 \times 10^{23}$ mol^{-1} is the Avogadro number and α is the static dipole polarizability of the atoms (or molecules) of the gas. This relation opens a possibility [10] of establishing pressure and/or temperature standards based on capacitance measurements of the dielectric constant, provided that α can be computed with sufficient accuracy, including small contributions such as mass-polarization, relativistic, and QED effects. Helium is evidently an ideal system for this purpose because the two-electron Schrödinger equation can be solved with very high precision. The published values of the infinite-nuclear-mass polarizability of helium have now reached an accuracy of 0.1 ppb [11]. This paper focuses on the relativistic contribution, for which existing literature data differ significantly [12–14].

After separating the center-of-mass motion, and neglecting the mass-polarization term $\hat{H}_{\mathrm{MP}} = -\frac{\hbar^2}{M}\nabla_1\nabla_2$, the nonrelativistic helium atom Hamiltonian becomes

$$\hat{H}_0 = -\frac{\hbar^2}{2\mu}(\nabla_1^2 + \nabla_2^2) - \frac{2e^2}{r_1} - \frac{2e^2}{r_2} + \frac{e^2}{r_{12}} \qquad (1)$$

and differs from the infinite-nuclear-mass Hamiltonian by containing the reduced mass $\mu = mM/(m + M)$ in place of the electron mass m. We will consider exclusively the ^{4}He isotope and assume that $M/m = 7294.2995$. If the problem is solved in the *reduced atomic units* [15] obtained by setting μ, e, and $\hbar$ equal to 1, the polarizability and the corrections to it are expressed in the units of $a_0^3(1 + m/M)^3$, where $a_0 = \hbar/me^2 = 0.529\,177\,21 \times 10^{-10}$ m is the Bohr radius. Similarly, the hyperpolarizability γ will be expressed in the units of $e^{-2}a_0^7(1 + m/M)^7$. Throughout this Letter we will always use these reduced atomic units and refer to them as atomic units (a.u.). The velocity of light c takes the value $c = 137.03600$ both in the reduced and conventional atomic units. The scaling of the units takes care of the main part of the finite-nuclear-mass effect. The remaining contribution can be accounted for by including the term $\hat{H}_{\mathrm{MP}}$ in the Hamiltonian.

The energy E_F of an atom placed in a static, uniform electric field of strength F can be expanded as $E_F = E_0 + F^2E_2 + F^4E_4 + \ldots$, where $E_2 = -\alpha/2$, $E_4 = -\gamma/24$, and E_0 is the energy for $F = 0$. Analogously, the wave function takes the form $\Phi_F = \Phi_0 + F\Phi_1 + F^2\Phi_2 + \ldots$. The functions Φ_k are defined by the Rayleigh-Schrödinger perturbation equations,

$$(\hat{H}_0 - E_0)\Phi_0 = 0, \qquad (2)$$

$$(\hat{H}_0 - E_0)\Phi_1 = -\hat{D}_x\Phi_0, \qquad (3)$$

$$(\hat{H}_0 - E_0)\Phi_2 = -\hat{D}_x\Phi_1 + \langle\Phi_1|\hat{D}_x|\Phi_0\rangle\Phi_0, \qquad (4)$$

and by a suitable normalization condition for Φ_F. $\hat{D}_x$ represents here the x component of the dipole moment operator $\hat{\boldsymbol{D}} = \boldsymbol{r}_1 + \boldsymbol{r}_2$. The exact, analytical solutions of Eqs. (2)–(4) are not known, but arbitrarily good approximations to Φ_k can be obtained variationally by minimizing the functionals [16]

$$J_0[\tilde{\Phi}_0] = \langle\tilde{\Phi}_0|\hat{H}_0|\tilde{\Phi}_0\rangle/\langle\tilde{\Phi}_0\,|\,\tilde{\Phi}_0\rangle, \qquad (5)$$

$$J_2[\tilde{\Phi}_1] = \langle\tilde{\Phi}_1|\hat{H}_0 - \bar{E}_0|\tilde{\Phi}_1\rangle + 2\langle\tilde{\Phi}_1|\hat{D}_x|\Psi_0\rangle, \qquad (6)$$

$$J_4[\tilde{\Phi}_2] = \langle\tilde{\Phi}_2|\hat{H}_0 - \bar{E}_0|\tilde{\Phi}_2\rangle + 2\langle\tilde{\Phi}_2|\hat{D}_x|\Psi_1\rangle - 2\bar{E}_2\langle\tilde{\Phi}_2\,|\,\Psi_0\rangle - \bar{E}_2\langle\Psi_1\,|\,\Psi_1\rangle. \qquad (7)$$

Ψ_k denotes here the approximation to Φ_k obtained by minimizing $J_{2k}[\tilde{\Phi}_k]$ with the trial function $\tilde{\Phi}_k$ expanded in a finite basis set. The corresponding minimum of $J_{2k}[\tilde{\Phi}_k]$

is denoted by $\bar{E}_{2k}$. In Eqs. (6) and (7), and later in this paper, it is assumed that all wave functions are real and that Ψ_0 is normalized, $\langle\Psi_0|\Psi_0\rangle = 1$. The functionals $J_2[\tilde{\Phi}_1]$ and $J_4[\tilde{\Phi}_2]$ provide rigorous upper bounds for E_2 and E_4, respectively, only if the functions Ψ_0 and Ψ_1 in Eqs. (6) and (7) are replaced by their exact counterparts Φ_0 and Φ_1. In practice, however, the use of high-quality approximations to these functions preserves the upper-bound property. When $\tilde{\Phi}_2$ is expanded in a basis with respect to which J_0 is stationary at Ψ_0, the functionals of Eqs. (5)–(7) correspond to the use of the (nonrelativistic) stationary perturbation theory [17].

The Breit-Pauli operator for singlet states of a two-electron atom interacting (or noninteracting) with a static electric field has the form [1] (in reduced units)

$$\hat{B} = -\frac{1}{8c^2}(\nabla_1^4 + \nabla_2^4) + \frac{\pi Z}{2c^2}[\delta(\boldsymbol{r}_1) + \delta(\boldsymbol{r}_2)] + \frac{\pi}{c^2}\delta(\boldsymbol{r}_{12}) + \frac{\nabla_1\nabla_2}{2c^2 r_{12}} + \frac{\boldsymbol{r}_{12}(\boldsymbol{r}_{12}\nabla_1)\nabla_2}{2c^2 r_{12}^3}, \tag{8}$$

where Z is the nuclear charge and $\delta(\boldsymbol{r})$ is the Dirac distribution. This expression results from the decoupling of "large" and "small" components in the Dirac-Coulomb-Breit equation to the leading order in c^{-2} [18]. $\hat{B}$ is a highly singular operator and can be used only to calculate expectation values. Its ∇_i^4 and $\delta(\boldsymbol{r}_i)$ terms probe mainly local properties of wave functions. This causes slow convergence when expectation values are computed with a series of approximate wave functions.

In the stationary variant [19] of the DPT, which is valid also for approximate zeroth-order wave functions, the leading-order relativistic correction is given by [5,19]

$$\Delta E_{\mathrm{DPT}}^{\mathrm{R}} = \langle\hat{B}\rangle + \frac{1}{2c^2}(\langle\hat{T}\hat{H}\rangle - \langle\hat{T}\rangle\langle\hat{H}\rangle), \tag{9}$$

where $\hat{T}$ is the kinetic energy operator, $\hat{H}$ is the nonrelativistic Hamiltonian, and the expectation values are computed with a wave function making $\langle\hat{H}\rangle$ stationary under some set of variations. If the exact eigenfunction of $\hat{H}$ is used, the second term in Eq. (9) vanishes and, in the lowest-order DPT becomes equivalent to the Breit-Pauli approach (note that DPT can be applied to all orders in c^{-2}, at least when using a self-consistent–type approach [18]). In practical, finite basis set calculations $\Delta E_{\mathrm{DPT}}^{\mathrm{R}}$ converges faster than $\langle\hat{B}\rangle$ to the infinite basis set limit [4]. One may also observe that the expectation value of the first two, singular terms in Eq. (8), and the first part ($\frac{1}{2}c^{-2}\langle\hat{T}\hat{H}\rangle$) of the correction term in Eq. (9), can be obtained [5] by a transformation of the DPT expression $-c^{-2}\langle\boldsymbol{\sigma}_1\nabla_1(\hat{V} + \hat{T}_2)\boldsymbol{\sigma}_1\nabla_1\rangle/2$, where $\boldsymbol{\sigma}_i$ and $\hat{T}_i$ are the vector of 2×2 Pauli spin matrices and the kinetic energy operator, respectively, for the ith electron, and $\hat{V} = \hat{H} - \hat{T}$. It is therefore clear that DPT involves global rather than local properties and should lead to faster convergence than the Breit-Pauli expression.

Inserting $\hat{H} = \hat{H}_0 + F\hat{D}_x$ into Eq. (9) and evaluating the expectation values with the electric-field-dependent wave function $\Psi_F = \Psi_0 + F\Psi_1 + F^2\Psi_2 + \ldots$ gives an expansion of $\Delta E_{\mathrm{DPT}}^{\mathrm{R}}$ in powers of F. Collecting terms of the order F^2, one obtains the following expression for the relativistic correction to the polarizability:

$$(\Delta\alpha)_{\mathrm{DPT}}^{\mathrm{R}} = (\Delta\alpha)_{\mathrm{BP}}^{\mathrm{R}} + \sum_{k=0}^{2}(\delta\alpha)_k^{\mathrm{R}}, \tag{10}$$

where

$$(\Delta\alpha)_{\mathrm{BP}}^{\mathrm{R}} = -2[\langle\Psi_1|\hat{B}|\Psi_1\rangle - \langle\Psi_0|\hat{B}|\Psi_0\rangle\langle\Psi_1|\Psi_1\rangle + 2\langle\Psi_2|\hat{B}|\Psi_0\rangle - 2\langle\Psi_0|\hat{B}|\Psi_0\rangle\langle\Psi_2|\Psi_0\rangle] \tag{11}$$

is the contribution from the Breit-Pauli operator and

$$-c^2(\delta\alpha)_0^{\mathrm{R}} = \langle\Psi_2|\hat{T}(\hat{H}_0 - \bar{E}_0)|\Psi_0\rangle - \langle\Psi_0|\hat{T}(\hat{H}_0 - \bar{E}_0)|\Psi_0\rangle[\langle\Psi_1|\Psi_1\rangle + 2\langle\Psi_2|\Psi_0\rangle] - 2\langle\Psi_2|(\hat{H}_0 - \bar{E}_0)|\Psi_0\rangle\langle\Psi_0|\hat{T}|\Psi_0\rangle, \tag{12}$$

$$-c^2(\delta\alpha)_1^{\mathrm{R}} = \langle\Psi_1|\hat{T}(\hat{H}_0 - \bar{E}_0)|\Psi_1\rangle + \langle\Psi_1|\hat{T}\hat{D}_x|\Psi_0\rangle, \tag{13}$$

$$-c^2(\delta\alpha)_2^{\mathrm{R}} = \langle\Psi_0|\hat{T}(\hat{H}_0 - \bar{E}_0)|\Psi_2\rangle + \langle\Psi_0|\hat{T}\hat{D}_x|\Psi_1\rangle - \langle\Psi_0|\hat{T}|\Psi_0\rangle\langle\Psi_1|\hat{D}_x|\Psi_0\rangle \tag{14}$$

are the correction terms predicted by the stationary DPT. Note that Ψ_1 and Ψ_2 in Eq. (11) give additive contributions, which can be denoted as $(\Delta\alpha)_{\mathrm{BP}}^{\mathrm{R1}}$ and $(\Delta\alpha)_{\mathrm{BP}}^{\mathrm{R2}}$. In the derivation of Eq. (13) we assumed that the linear algebraic equation resulting from the stationarity condition $\delta J_2 = 0$ is solved exactly. It can be seen that $(\delta\alpha)_k^{\mathrm{R}}$ vanish if the functions Ψ_k are replaced by their exact counterparts Φ_k (which rationalizes our grouping of the terms). Thus, $(\delta\alpha)_k^{\mathrm{R}}$ tests the accuracy of the solution of the kth-order equation. One may also note that the last term in Eq. (12) vanishes when Ψ_2 is expanded in a basis with respect to which J_0 is stationary at Ψ_0. Our basis sets do not fulfill this condition so this term is kept in Eq. (12).

The functionals (5)–(7) were minimized one after another by expanding the trial functions $\tilde{\Phi}_0$, $\tilde{\Phi}_1$, and $\tilde{\Phi}_2$ in the basis of Gaussian-type geminals [20]:

$$\tilde{\Phi}_k = \hat{P}_k(1 + \hat{P}_{12})\sum_{i=1}^{N_k} c_i^{(k)}\phi_i^{(k)}, \tag{15}$$

$$\phi_i^{(k)} = \exp(-\alpha_{1i}^{(k)}|\boldsymbol{r}_1 - \boldsymbol{A}_i^{(k)}|^2 - \alpha_{2i}^{(k)}|\boldsymbol{r}_2 - \boldsymbol{B}_i^{(k)}|^2 - \beta_i^{(k)}|\boldsymbol{r}_1 - \boldsymbol{r}_2|^2), \tag{16}$$

where $\hat{P}_{12}$ denotes the permutation of the spatial electronic coordinates $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$, $\hat{P}_0 = 1$, $\hat{P}_1 = 1 - \hat{I}$, and $\hat{P}_2 = 1 + \hat{I}$, with $\hat{I}$ being the inversion through the point

TABLE I. Convergence of the nonrelativistic polarizability α (without the mass-polarization correction) and the Ψ_1-dependent relativistic contributions [see Eqs. (11) and (13) and the text below Eq. (14)] with the expansion length N_1 for Ψ_1. Ψ_0 was always expanded in the 1200-term basis. All quantities are in reduced atomic units.

N_1	α	$(\Delta\alpha)^{\rm R1}_{\rm BP} \times 10^6$	$(\delta\alpha)^{\rm R}_1 \times 10^6$
300	1.383 192 170 07	44.6350	0.00024
424	1.383 192 173 33	44.6386	0.00039
600	1.383 192 174 00	44.6404	0.00047
848	1.383 192 174 17	44.6417	0.00033
Ref. [11]	1.383 192 174 44(5)		
Ref. [21]	1.383 192 174 455(1)		

(0,0,0). The centers $\boldsymbol{A}_i^{(0)}$ and $\boldsymbol{B}_i^{(0)}$ have coordinates (0,0,0), while $\boldsymbol{A}_i^{(k)}$ and $\boldsymbol{B}_i^{(k)}$ $(k = 1, 2)$ are constrained to the x axis. The function $\tilde{\Phi}_0$ has the $S^{\rm e}$ symmetry under the operations of the $O(3)$ group, while $\tilde{\Phi}_1$ and $\tilde{\Phi}_2$ transform according to the Σ_u^+ and Σ_g^+ representations, respectively, of the $D_{\infty h}$ group. Since $D_{\infty h}$ is a subgroup of $O(3)$, the function $\tilde{\Phi}_1$ can become arbitrarily close (when N_0 and N_1 tend to infinity) to the exact function Φ_1, which is of $P^{\rm o}$ symmetry. Analogously, when N_0, N_1, and N_2 increase to infinity, $\tilde{\Phi}_2$ can approximate arbitrarily well the exact Φ_2, which is a mixture of $S^{\rm e}$ and $D^{\rm e}$ symmetries. The only practical problem with this approach is a numerical cancellation of significant digits caused by the action of the operator $P_1 = 1 - \hat{I}$ when both $a_{ix}^{(1)}$ and $b_{ix}^{(1)}$ are very close to zero, which forced us to reject some functions in the optimization process.

The minimization was done by variations of the nonlinear parameters $\alpha_{1i}^{(k)}$, $\alpha_{2i}^{(k)}$, $\beta_i^{(k)}$, $\boldsymbol{A}_i^{(k)}$, and $\boldsymbol{B}_i^{(k)}$ in functions (16) (see Ref. [20] for details of the algorithm), each variation followed by the solution of an appropriate system of N_k linear equations for the $c_i^{(k)}$ coefficients. All the time-consuming optimizations of the variational parameters were done in the standard 64-bit precision, but the final results were recomputed using 128-bit precision.

As the zeroth-order function Ψ_0, we used the 1200-term expansion from Ref. [4], which yields 11 exact energy digits and is the most accurate Gaussian function for the helium atom available today. We tested different expansion lengths for Ψ_1 and Ψ_2. In Table I we present the convergence of the nonrelativistic polarizability α and Ψ_1-dependent relativistic contributions $(\Delta\alpha)^{\rm R1}_{\rm BP}$ and $(\delta\alpha)^{\rm R}_1$ with the expansion length for Ψ_1. The convergence of α is smooth and the 10 significant digits obtained with the 848-term Gaussian basis is more than satisfactory for our purposes. It should be noted, however, that our values of α appear to converge to a limit which is by about 2×10^{-10} a.u. smaller than the best literature estimate [11,21]. This is a result of the rejection of some functions in the optimization process as described in the symmetry discussion below Eq. (16). The Ψ_1 contribution to $(\Delta\alpha)^{\rm R}_{\rm BP}$ is converged to at least 10^{-8} a.u. and has—within this accuracy—identical value with the DPT prediction, since the DPT correction term $(\delta\alpha)^{\rm R}_1$ is in all cases smaller than 10^{-9} a.u. The value of $(\Delta\alpha)^{\rm R1}_{\rm BP}$ equal to 44.64 agrees reasonably well with Weinhold's [12] result of 46.97 (Weinhold did not consider the Ψ_2 contribution to $(\Delta\alpha)^{\rm R}_{\rm BP}$) . Note, however, that the sign of $(\Delta\alpha)^{\rm R1}_{\rm BP}$ given in Ref. [12] is incorrect.

The convergence of the components which depend on Ψ_2 is shown in Table II. Evidently, it is much harder to obtain an accurate Ψ_2 than Ψ_1 (probably because it has to be converged in both S^e and D^e subspaces): despite using expansions as large as $N_2 = 2400$, the nonrelativistic value of the second hyperpolarizability, γ, is converged with respect to N_2 to only 9 digits. The values of $(\delta\alpha)^{\rm R}_0$ turn out to be strongly dominated by the first term in Eq. (12). The values of $(\Delta\alpha)^{\rm R}_{\rm BP}$ and $(\Delta\alpha)^{\rm R}_{\rm DPT}$ were computed using our most accurate 848-term representation for Ψ_1. While the convergence of $(\Delta\alpha)^{\rm R}_{\rm BP}$ is slow and obscured by strong oscillations, the DPT results converge fairly smoothly and monotonically to an extrapolated value of

$$(\Delta\alpha)^{\rm R}_{\rm DPT} = -0.000\,080\,34(2)\ \text{a.u.}, \tag{17}$$

where the error bar estimation results from testing various

TABLE II. The nonrelativistic hyperpolarizability γ (without the mass-polarization correction), the total relativistic corrections to the polarizability $(\Delta\alpha)^{\rm R}_{\rm BP}$ and $(\Delta\alpha)^{\rm R}_{\rm DPT}$, and the Ψ_2-dependent contributions to $(\Delta\alpha)^{\rm R}_{\rm DPT} - (\Delta\alpha)^{\rm R}_{\rm BP}$ as functions of the length N_2 of the expansion for Ψ_2. Ψ_0 was always expanded in the 1200-term basis and Ψ_1 in the 848-term basis. All quantities are expressed in reduced atomic units and (except for γ) multiplied by 10^6.

N_2	γ	$(\Delta\alpha)^{\rm R}_{\rm BP}$ Eq. (11)	$(\delta\alpha)^{\rm R}_0$ Eq. (12)	$(\delta\alpha)^{\rm R}_2$ Eq. (14)	$(\Delta\alpha)^{\rm R}_{\rm DPT}$ Eq. (10)
300	43.104 202 421	−81.7261	0.0009	1.2937	−80.4312
424	43.104 221 524	−80.2421	0.0027	−0.1627	−80.4017
600	43.104 225 756	−80.3901	0.0057	−0.0023	−80.3863
848	43.104 227 219	−80.6143	0.0057	0.2247	−80.3835
1200	43.104 227 560	−80.6311	0.0049	0.2550	−80.3709
1697	43.104 227 652	−80.3780	0.0046	0.0070	−80.3661
2400	43.104 227 696	−80.2940	0.0043	−0.0693	−80.3586
Ref. [22]	43.104				

reasonable least-squares fits assuming $N^{-\kappa}$ or $\exp(-aN^{\kappa})$ convergence. This result is in excellent agreement with the very recent value of $-0.000\,080\,358(27)$ a.u. obtained by Pachucki and Sapirstein [21] and slightly but significantly disagrees with Bhatia and Drachman's value of $-0.000\,080\,029$ a.u. [14]. The configuration interaction result of Ref. [13], $(\Delta\alpha)^{\mathrm{R}} = -0.000\,076\,5$ a.u., has to be regarded as less accurate, evidently due to the use of insufficiently flexible wave functions. It should be stressed that our calculation, unlike the others discussed here, was performed with a general basis set, not specifically adapted to the atomic case, which means that similar quality calculations could be done for molecules.

We have checked how the use of the approximate wave functions Ψ_0 and Ψ_1 in Eqs. (6) and (7) influences our final results. For this purpose, we generated a double series of 2400-term Ψ_2 functions, which where computed using different expansion lengths for Ψ_0 ($N_0 = 150, 300, 600, 1200$) and for Ψ_1 ($N_1 = 300, 424, 600, 848$). The extrapolation to the limit $N_0 \rightarrow \infty$ changed nothing in our results, which proves that the 1200-term expansion for Ψ_0 is sufficiently saturated. Similarly, the relativistic polarizability corrections were fairly insensitive to the value of N_1. However, the values of γ obtained with fixed values of $N_0 = 1200$ and $N_2 = 2400$ and with N_1 equal to 300, 424, 600, 848 are 43.104 228 67, 43.104 228 12, 43.104 227 88, and 43.104 227 70, respectively. Thus, not all digits which appear converged in the second column of Table II can be regarded as certain, and our recommended value for γ is 43.104 227(1), which is 3 orders of magnitude more accurate than the best previous literature value [22].

We also calculated the mass-polarization effect on the polarizability and hyperpolarizability by solving Eqs. (2)–(4) with the $\hat{H}_{\mathrm{MP}}$ operator explicitly included in $\hat{H}_0$. The differences between the values with and without the $\hat{H}_{\mathrm{MP}}$ term are almost independent of the expansion lengths of Ψ_1 and Ψ_2 and amount to $(\Delta\alpha)^{\mathrm{MP}} = 0.000\,048\,834\,5$ a.u. and $(\Delta\gamma)^{\mathrm{MP}} = 0.005\,237\,173$ a.u. The functions Ψ_k containing the effect of $\hat{H}_{\mathrm{MP}}$ could also, in principle, be used to evaluate the coupling between this operator and the relativistic effects. However, additional finite-nuclear-mass effects on the relativistic correction (apart from the included trivial effect resulting from using reduced atomic units) exist [23] and are much more difficult to compute. These nontrivial contributions, known as the nuclear recoil corrections, as well as the trivial ones, are of the order of $c^{-2}M^{-1}$ and can enter the total polarizability value, at most, at the eighth place after the decimal point, i.e., are comparable to the convergence error of our value of $(\Delta\alpha)^{\mathrm{R}}_{\mathrm{DPT}}$. Their inclusion would be desirable if the much larger QED corrections (of the order of $c^{-3}\ln c$) were known with sufficient accuracy.

Our value of $(\Delta\alpha)^{\mathrm{R}}_{\mathrm{DPT}}$, given by Eq. (17), can be combined with the nonrelativistic polarizability from Table I, with the mass-polarization correction, $(\Delta\alpha)^{\mathrm{MP}}$, given above, and with the estimated QED correction [21], $(\Delta\alpha)^{\mathrm{QED}} = 0.000\,030(2)$ a.u., to give the total value $\alpha = 1.383\,191(2)$ a.u. This yields our recommended value of the molar polarizability of ^{4}He,

$$A_{\varepsilon} = 0.517\,254(1)\ \mathrm{cm}^3\,\mathrm{mol}^{-1}, \tag{18}$$

where the uncertainty results almost completely from an approximate calculation of QED effects in Ref. [21].

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