

Radiative Corrections to the Polarizability of Helium

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The complete α^3 QED correction to the helium atom polarizability is computed assuming an infinite nuclear mass and found to be equal to 0.000 030 666(3) a.u., with the contribution from the electric-field dependence of the Bethe logarithm amounting to 0.000 000 193(2) a.u. After including the α^2 and α^3 corrections for the nuclear recoil and the leading part of the α^4 QED correction, we find that the molar polarizability of ^4He is 0.517 254 19(9)(4) cm^3/mol . The first of the two error bounds is dominated by the uncertainty of α^4 and higher-order QED corrections and the second reflects the uncertainty of the Avogadro constant.

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The well-known statistical mechanics relation between the pressure of a gas, its temperature, dielectric constant, and molar polarizability [1–3] opens up a possibility of establishing pressure and/or temperature standards based on capacitance measurements of the dielectric constant. The uncertainty of the determined pressure would then depend on the uncertainty of the molar polarizability of the gas and of its virial coefficients. For helium these uncertainties can be reduced to an acceptable level by including relativistic and quantum electrodynamics (QED) corrections to the polarizability and by recomputing the pair potential for helium using the state of the art perturbation theory and supermolecular techniques [4].

The nonrelativistic value of the polarizability of helium is known to about 1 ppb, judging from the convergence of recent *ab initio* calculations [5–7]. The relativistic correction of the order of α^2 , where $\alpha = 1/137.036\,00$ is the fine structure constant, has recently been calculated by Pachucki and Sapirstein [6] and by Cencek *et al.* [7]. These calculations resolved the discrepancy between earlier results of Johnson and Cheng [8] and of Bhatia and Drachman [5] showing that the latter result is much more accurate than the former one. Pachucki and Sapirstein estimated also the QED correction to the helium polarizability by calculating accurately all contributions of the order of α^3 , except those containing the electric-field derivative of the so-called Bethe logarithm [9]. Their value, despite being of the order of α^3 , represents about 40% of the α^2 relativistic correction. Pachucki and Sapirstein estimated that the error of their result for He atom polarizability is of the order of 2 ppm.

In the present communication we report accurate calculations of the α^3 QED corrections to the helium polarizability including an accurate treatment of the contribution resulting from the field dependence of the Bethe logarithm. To allow for the rapid variation of the perturbed wave functions corresponding to high momenta of the exchanged photon we used the basis set of exponentially correlated Slater functions with fully optimized

exponents. The α^3 terms computed previously by Pachucki and Sapirstein were also calculated to verify their accuracy. We shall also present an estimation of the $\alpha^2 m_e/M_{\text{He}}$ relativistic recoil correction as well as the $\alpha^3 m_e/M_{\text{He}}$ and α^4 QED corrections.

The nonrelativistic dipole polarizability α_d of helium can be calculated directly from its definition using the second-order perturbation theory formula:

$$\alpha_d = \frac{2}{3} \langle \psi_0 | \mathbf{r} \mathcal{R}_0 \mathbf{r} | \psi_0 \rangle, \quad (1)$$

where ψ_0 is the ground-state wave function, \mathcal{R}_0 is the reduced resolvent

$$\mathcal{R}_0 = Q(QH_0 - E_0)^{-1}$$

of the nonrelativistic helium atom Hamiltonian

$$H_0 = \frac{\mathbf{p}_1^2}{2} + \frac{\mathbf{p}_2^2}{2} - \frac{2}{r_1} - \frac{2}{r_2} + \frac{1}{r_{12}}, \quad (2)$$

E_0 is the ground-state energy, $Q = 1 - |\psi_0\rangle\langle\psi_0|$, and $\mathbf{r} = \mathbf{r}_1 + \mathbf{r}_2$, with \mathbf{r}_i pointing from the nucleus to the electron i . In Eq. (2) we assumed an infinite nuclear mass, although including the nuclear motion does not present any difficulties at the nonrelativistic level. In Eq. (1) and further on, atomic units are used, i.e., $\hbar = 1$, $e = 1$, and $m_e = 1$.

When the Hamiltonian H_0 is modified by a perturbation δH , the corresponding change in the polarizability is given by (keeping only terms linear in δH)

$$\begin{aligned} \delta\alpha_d = & -\frac{4}{3} \langle \psi_0 | \mathbf{r} \mathcal{R}_0 \mathbf{r} \mathcal{R}_0 \delta H | \psi_0 \rangle \\ & - \frac{2}{3} \langle \psi_0 | \mathbf{r} \mathcal{R}_0 (\delta H - \langle \delta H \rangle) \mathcal{R}_0 \mathbf{r} | \psi_0 \rangle. \end{aligned} \quad (3)$$

This formula allows us to calculate the lowest order relativistic corrections to the polarizability, corrections resulting from finite mass of the nucleus and, more generally, all corrections which can be obtained by an

electric-field differentiation of an expectation value of δH .

When the nuclear recoil is neglected, the α^3 QED correction to the energy of the ground state of helium is given by the formula [10,11]

$$E_{\text{QED}} = \alpha^3 \frac{8}{3} \left(\frac{19}{30} - 2 \ln \alpha - \ln k_0 \right) \langle \psi_0 | \delta^3(\mathbf{r}_1) + \delta^3(\mathbf{r}_2) | \psi_0 \rangle \\ + \alpha^3 \left(\frac{164}{15} + \frac{14}{3} \ln \alpha \right) \langle \psi_0 | \delta^3(\mathbf{r}_{12}) | \psi_0 \rangle \\ - \alpha^3 \frac{7}{6\pi} \langle \psi_0 | P(r_{12}^{-3}) | \psi_0 \rangle. \quad (4)$$

The distribution $P(r_{12}^{-3})$ is defined by

$$\langle \psi_0 | P(r_{12}^{-3}) | \psi_0 \rangle = \lim_{a \rightarrow 0} \langle \psi_0 | \theta(r_{12} - a) r_{12}^{-3} \\ + 4\pi(\gamma + \ln a) \delta^3(\mathbf{r}_{12}) | \psi_0 \rangle, \quad (5)$$

where γ is the Euler constant and $\theta(x)$ is the Heaviside step function. The Bethe logarithm, $\ln k_0$, is a quotient,

$$\ln k_0 = \frac{\langle \psi_0 | \mathbf{p} (H_0 - E_0) \ln[2(H_0 - E_0)] \mathbf{p} | \psi_0 \rangle}{\langle \psi_0 | \mathbf{p} (H_0 - E_0) \mathbf{p} | \psi_0 \rangle}, \quad (6)$$

where $\mathbf{p} = \mathbf{p}_1 + \mathbf{p}_2$. The denominator in Eq. (6) can be simplified to the form $\mathcal{D} = 4\pi \langle \psi_0 | \delta^3(\mathbf{r}_1) + \delta^3(\mathbf{r}_2) | \psi_0 \rangle$.

The derivation of Eq. (4) can be repeated without much difficulty for the case of helium atom in external static electric field \mathcal{E} . The only difference would be a replacement of H_0 by $H = H_0 + \mathcal{E}r$. E_0 and ψ_0 would then be replaced by E and ψ , the ground state energy and wave function of H ,

$$E = E_0 - \frac{1}{2} \alpha_d \mathcal{E}^2 + \mathcal{O}(\mathcal{E}^4), \quad (7)$$

$$\psi = \psi_0 - \mathcal{R}_0 \mathcal{E} r \psi_0 + \mathcal{R}_0 \mathcal{E} r \mathcal{R}_0 \mathcal{E} r \psi_0 \\ - \frac{1}{2} \langle \psi_0 | \mathcal{E} r \mathcal{R}_0^2 \mathcal{E} r | \psi_0 \rangle \psi_0 + \mathcal{O}(\mathcal{E}^3). \quad (8)$$

If the Bethe logarithm were independent of the electric field, it would be possible to calculate the α^3 correction to the polarizability using Eq. (3). Corrections resulting from $\delta^3(\mathbf{r}_i)$ and $\delta^3(\mathbf{r}_{12})$ have already been calculated in the work on the relativistic effects [6,7]. The treatment of the distribution $P(r_{12}^{-3})$ causes no significant difficulties [6]. We found that the contribution of the last term in Eq. (4) to the polarizability of helium amounts to $0.37161(1) \times 10^{-6}$, in agreement with the results reported in Ref. [6].

The main computational challenge is the electric-field dependence of the Bethe logarithm. It introduces an additional correction to the polarizability,

$$\delta \alpha_d = \alpha^3 \frac{8}{3} \langle \psi_0 | \delta^3(\mathbf{r}_1) + \delta^3(\mathbf{r}_2) | \psi_0 \rangle \partial_{\mathcal{E}}^2 \ln k_0, \quad (9)$$

where the electric-field derivative is taken at $\mathcal{E} = 0$. The computation of the numerator in Eq. (6), and its second derivative over the electric field, is rather involved. The

main problem is a large contribution coming from intermediate states of very high energies. As shown by Drake and Goldman [12], it is possible to calculate $\ln k_0$ directly from formula (6), but to obtain satisfactory precision one should use basis functions varying rapidly at very small distances. We expect this problem to be even more severe in the calculation of the second derivative of the numerator, and we decided to use the integral representation of the Bethe logarithm,

$$\ln k_0 = \lim_{\Lambda \rightarrow \infty} \left[-\frac{\langle \mathbf{p}^2 \rangle}{\mathcal{D}} \Lambda + \ln(2\Lambda) + \int_0^\Lambda \omega \frac{J(\omega)}{\mathcal{D}} d\omega \right], \quad (10)$$

where $\langle \mathbf{p}^2 \rangle = \langle \psi | \mathbf{p}^2 | \psi \rangle$ and for $\omega > 0$:

$$J(\omega) = \langle \psi | \mathbf{p} (H - E + \omega)^{-1} \mathbf{p} | \psi \rangle. \quad (11)$$

This formula follows directly from the calculation of the contribution of single transverse photon exchange to the Lamb shift. The high-energy region, corresponding to high frequency of the exchanged photon, can be included with the help of the asymptotic formula [13]

$$\frac{J(\omega)}{\mathcal{D}} = \frac{1}{\omega} \frac{\langle \mathbf{p}^2 \rangle}{\mathcal{D}} - \frac{1}{\omega^2} + \frac{4}{\omega^3} \left[\sqrt{2\omega} - 2 \ln \omega + C_3 + C_4 \frac{1}{\sqrt{\omega}} \right. \\ \left. + C_5 \frac{\ln \omega}{\omega} + C_6 \frac{1}{\omega} + \dots \right]. \quad (12)$$

The coefficients C_i in this asymptotic expansion can be determined by fitting to known values of $J(\omega)/\mathcal{D}$. However, C_3 can be calculated directly. As shown by Schwartz [13], and confirmed by Forrey and Hill [14], C_3 can be written as

$$C_3 = \frac{1}{\rho(0)} \left[\left(\frac{1}{2} \ln 2 - \frac{1}{2} - \gamma \right) \frac{d\rho(r)}{dr} \Big|_{r=0} \right. \\ \left. - \int_0^\infty dr \ln r \frac{d^2 \rho(r)}{dr^2} + 2 \langle \psi | \mathbf{r}_1 \mathbf{r}_2 (r_1 r_2)^{-3} | \psi \rangle \right], \quad (13)$$

where $\rho(r)$ is the radial electron density. We found that $C_3 = 5.00078(3)$ at zero field, in fair agreement with the value 4.988(1) obtained from the fitting [15]. Although a formula for C_4 was derived [14], it is far more complex than the one for C_3 and we decided not to use it, but obtain C_4 , together with C_5 and C_6 for which analytic formulas are not known, by fitting.

Denoting the resolvent $(H_0 - E_0 + \omega)^{-1}$ by $\mathcal{R}(\omega)$ and the Cartesian coordinates of vectors \mathbf{r} and \mathbf{p} by r^i and p^i , $i = x, y, z$, we can express the second-order electric-field derivative of $J(\omega)$ in following compact form:

$$\begin{aligned} \partial_{\mathcal{E}}^2 J(\omega) = & \frac{2}{3} [2\langle \psi_0 | r^i \mathcal{R}_0 r^i \mathcal{R}_0 p^j \mathcal{R}(\omega) p^j | \psi_0 \rangle + 2\langle \psi_0 | r^i \mathcal{R}_0 p^j \mathcal{R}(\omega) r^i \mathcal{R}(\omega) p^j | \psi_0 \rangle + \langle \psi_0 | r^i \mathcal{R}_0 p^j \mathcal{R}(\omega) p^j \mathcal{R}_0 r^i | \psi_0 \rangle \\ & - \langle \psi_0 | p^j \mathcal{R}(\omega) p^j | \psi_0 \rangle \langle \psi_0 | r^i \mathcal{R}_0^2 r^i | \psi_0 \rangle + \langle \psi_0 | p^j \mathcal{R}(\omega) r^i \mathcal{R}(\omega) r^i \mathcal{R}(\omega) p^j | \psi_0 \rangle \\ & - \langle \psi_0 | r^i \mathcal{R}_0 r^i | \psi_0 \rangle \langle \psi_0 | p^j \mathcal{R}(\omega)^2 p^j | \psi_0 \rangle], \end{aligned} \quad (14)$$

where the summation over repeated indices is implicit. One can verify that the right-hand side (rhs) of Eq. (14) can be written as a sum of integrals involving two first-order wave functions, $\psi_r^i \equiv \mathcal{R}_0 r^i \psi_0$ and $\psi_p^j \equiv \mathcal{R}(\omega) p^j \psi_0$, and three second-order ones, $\mathcal{R}_0 r^i \psi_r^j$, $\mathcal{R}(\omega) p^j \psi_r^i$, and $\mathcal{R}(\omega) r^i \psi_p^j$.

To evaluate the rhs of Eq. (14) and all other matrix elements needed in this work, we used the basis set of exponentially correlated Slater-type functions:

$$Y(\mathbf{r}_1, \mathbf{r}_2) e^{-\alpha_n r_1 - \beta_n r_2 - \gamma_n r_{12}} + (\mathbf{r}_1 \leftrightarrow \mathbf{r}_2), \quad (15)$$

where $Y(\mathbf{r}_1, \mathbf{r}_2) = 1$ for the ground state and for other S states that appear in our calculations. For the P states of vector and pseudovector symmetries we set $Y(\mathbf{r}_1, \mathbf{r}_2) = \mathbf{r}_1$ and $Y(\mathbf{r}_1, \mathbf{r}_2) = \mathbf{r}_1 \times \mathbf{r}_2$, respectively. The basis sets for D states consisted of two parts with $Y(\mathbf{r}_1, \mathbf{r}_2) = r_1^i r_1^j - \frac{1}{3} r_1^2 \delta^{ij}$ and $Y(\mathbf{r}_1, \mathbf{r}_2) = r_{12}^i r_{12}^j - \frac{1}{3} r_{12}^2 \delta^{ij}$. All the nonlinear parameters $\{\alpha_n, \beta_n, \gamma_n\}$ were optimized variationally. In the case of ground state we minimized the Rayleigh quotient: $\langle \tilde{\psi}_0 | H_0 | \tilde{\psi}_0 \rangle / \langle \tilde{\psi}_0 | \tilde{\psi}_0 \rangle$. To find the optimal pa-

rameters of first- and second-order wave functions needed to evaluate the rhs of Eq. (14) we used the functional

$$\mathcal{F}[\tilde{\psi}] = \langle \tilde{\psi} | H_0 - E_0 + \omega | \tilde{\psi} \rangle + 2\langle \tilde{\psi}_1 | f \rangle, \quad (16)$$

where the function f is equal to $r^i \psi_0$ or $p^j \psi_0$ for the first-order wave functions and to $r^i \psi_r^j$, $p^j \psi_r^i$, or $r^i \psi_p^j$ for the second-order ones. For the expansion of the zeroth-, first-, and second-order wave functions we used N_0 , N_1 , and N_2 basis functions, respectively.

Note that $\partial_{\mathcal{E}}^2 J(0) = 0$, but the individual terms in Eq. (14) are finite or even singular at $\omega = 0$. The effective numerical cancellation of this singularity was a good accuracy check of our method to evaluate $\partial_{\mathcal{E}}^2 J(\omega)$.

Almost all of the integrals encountered in our work can be calculated from ‘‘master integrals’’ given in [16] by repeated differentiations over parameters α , β , and γ . Some additional work was required to calculate the C_3 coefficient, Eq. (13). In this case the following integral, involving the dilogarithm function $\text{Li}_2(z)$ [17], was used:

$$\begin{aligned} \frac{1}{16\pi^2} \int \mathbf{r}_1 \mathbf{r}_2 \frac{e^{-\alpha r_1 - \beta r_2 - \gamma r_{12}}}{r_1^3 r_2^3 r_{12}} d^3 \mathbf{r}_1 d^3 \mathbf{r}_2 = & \frac{1}{\gamma} \left[1 + \frac{\alpha}{\gamma} \ln \frac{\alpha + \beta}{\alpha + \gamma} + \frac{\beta}{\gamma} \ln \frac{\beta + \alpha}{\beta + \gamma} \right] \\ & - \frac{\alpha \beta}{2\gamma^3} \left[\frac{\pi^2}{6} + \frac{1}{2} \ln^2 \left(\frac{\gamma + \alpha}{\gamma + \beta} \right) + \text{Li}_2 \left(1 - \frac{\alpha + \beta}{\alpha + \gamma} \right) + \text{Li}_2 \left(1 - \frac{\beta + \alpha}{\beta + \gamma} \right) \right]. \end{aligned} \quad (17)$$

The integration over ω was done in a similar way as in Ref. [18]. The integration interval $[0, \infty]$ was split into intervals $[0, 1]$ and $[1, \infty]$, and in the second interval the integration variable was changed to $\zeta = \omega^{-1/2}$ transforming this interval into $[0, 1]$. We used the Gauss-Lobato quadratures in both parts. In comparison to Gauss-Legendre integration, it allows us to reduce the number of integration nodes without increasing the error of integration. The Gauss quadratures are, by definition, exact for polynomials up to certain order. In the calculation of $\ln k_0$ without external electric field, the presence of the logarithmic term in Eq. (12) is the main factor limiting the precision of the quadrature. Fortunately, the coefficient of the leading logarithmic term is field inde-

pendent and vanishes when $\partial_{\mathcal{E}}^2 J(\omega)$ is evaluated, which results in a faster convergence of the integral over ω . For $\omega > 500$, the asymptotic formula of Eq. (12) was used. We found that $\partial_{\mathcal{E}}^2 C_3 = -0.0805(1)$ and used this value in our calculations. To accurately include the contribution from $1 \ll \omega < 500$, we need a highly accurate value of the second derivative of $\langle \mathbf{p}^2 \rangle$ over the electric field. Using the virial theorem we found that $\partial_{\mathcal{E}}^2 \langle \mathbf{p}^2 \rangle = -6\alpha_d + 2\partial_{\mathcal{E}}^2 \langle \psi | \mathbf{p}_1 \mathbf{p}_2 | \psi \rangle = -9.01198117(1)$.

The convergence patterns of our calculation are displayed in Tables I, II, and III. It is seen that the convergence of $\partial_{\mathcal{E}}^2 \ln k_0$ is rather slow. We conservatively estimate that

$$\partial_{\mathcal{E}}^2 \ln k_0 = 0.0512(4). \quad (18)$$

TABLE I. Convergence of the $\ln k_0$ and $\partial_{\mathcal{E}}^2 \ln k_0$ with N_1 ; $N_0 = 256$, $N_2 = 512$.

Basis size	$\ln k_0$	$\partial_{\mathcal{E}}^2 \ln k_0$
64	4.369 459 157	0.036 600 547
128	4.370 116 219	0.050 351 859
256	4.370 158 946	0.051 186 969
512	4.370 160 105	0.051 412 345
Ref. [15]	4.370 159 (2)	
Ref. [19]	4.370 160 218(3)	

TABLE II. Convergence of the $\ln k_0$ and $\partial_{\mathcal{E}}^2 \ln k_0$ with increasing N_0 , and fixed $N_1 = 512$ and $N_2 = 512$.

Basis size	$\ln k_0$	$\partial_{\mathcal{E}}^2 \ln k_0$
64	4.370 296 134	0.071 617 005
128	4.370 174 403	0.052 448 318
256	4.370 160 105	0.051 412 345
512	4.370 160 457	0.051 437 640

TABLE III. Convergence of $J(\omega)$ (for $\omega = 1$ and $\omega = 503$) and $\partial_{\xi}^2 \ln k_0$ with N_2 ; $N_0 = 256$, $N_1 = 512$.

Basis size	$\partial_{\xi}^2 J(1)$	$\partial_{\xi}^2 J(503)$	$\partial_{\xi}^2 \ln k_0$
128	-2.473 881 216	-0.019 923 382	0.093 716 117
256	-2.473 879 822	-0.019 923 112	0.052 082 503
512	-2.473 879 745	-0.019 923 099	0.051 412 345
1024	-2.473 879 739	-0.019 923 097	0.051 243 409

The results of our calculations are summarized in Table IV. The nonrelativistic part includes the effect of motion of the nucleus obtained by adding the mass polarization term $(\mu/M_{\text{He}})\mathbf{p}_1 \times \mathbf{p}_2$ to the Hamiltonian of Eq. (2) and working in reduced atomic units, $\hbar = 1$, $e = 1$, and $\mu = 1$, where $\mu = m_e M_{\text{He}}/(m_e + M_{\text{He}})$.

We still need an estimate of the α^2 relativistic [20] and α^3 QED [21] recoil corrections. An accurate calculation of these contributions is not very difficult and we plan to perform it in the future. For the present work it is sufficient to use our preliminary estimate of the $\alpha^2 m_e/M_{\text{He}}$ correction equal to $-0.07(2) \times 10^{-6}$ a.u. The recoil correction of the order $\alpha^3 m_e/M_{\text{He}}$ is estimated to be smaller than 0.05×10^{-6} a.u.

The calculation of the α^4 correction to the ionization energy of helium is extremely complicated and was completed only very recently [16,19]. It can be expected that an analogous calculation for the α^4 corrections to the polarizability will be even more difficult and was not attempted. It is possible, however, to estimate the α^4 contribution by computing its dominant part given by the one-loop binding correction to the Lamb shift [16]:

$$4\pi\alpha^4 (427/96 - 2 \ln 2) \langle \psi_0 | \delta^3(\mathbf{r}_1) + \delta^3(\mathbf{r}_2) | \psi_0 \rangle. \quad (19)$$

This approximation works very well for the ionization energy of helium and reproduces the total α^4 contribution with an error of less than 10% [16,19]. If Eq. (19) is used to calculate the correction to the polarizability, one obtains 0.56×10^{-6} a.u. as an estimate of the α^4 contribu-

TABLE IV. Contributions to the polarizability of ${}^4\text{He}$ in atomic units $\times 10^{-6}$.

Nonrelativistic approximation	1 383 809.986...
α^2 relativistic corrections	-80.35(2)
Ref. [6]	-80.358(27)
Ref. [7]	-80.34(2)
α^3 QED without $\partial_{\xi}^2 \ln k_0$ contribution	30.473(1)
Ref. [6]	30.474(1)
α^3 QED contribution from $\partial_{\xi}^2 \ln k_0$	0.193(2)
α^4 , $\alpha^2 m_e/M_{\text{He}}$, and $\alpha^3 m_e/M_{\text{He}}$ terms	0.49(23)
Total	1 383 760.79(23)

tion. The error of this approximation is assumed very conservatively as 40%, which translates into a 0.16 ppm error in the total molar polarizability of helium.

Using the data from Table IV results in $0.517\,254\,19(9)(4)$ cm³/mol as our recommended value of the molar polarizability of helium. The first of the two error bounds is of theoretical origin and the second reflects the uncertainty of the Avogadro constant [1]. The overall error of our calculations, amounting to 0.2 ppm, is an order of magnitude smaller than that of previous theoretical determinations of the molar polarizability of helium.

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