

**Lithium electric dipole polarizability**

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The electric dipole polarizability of the lithium atom in the ground state is calculated including relativistic and quantum electrodynamics corrections. The obtained result  $\alpha_E = 164.0740(5)$  a.u. is in good agreement with the less accurate experimental value of  $164.19(1.08)$  a.u. The small uncertainty of about 3 parts per  $10^6$  comes from the approximate treatment of quantum electrodynamics corrections. Our theoretical result can be considered as a benchmark for more general atomic structure methods and may serve as a reference value for the relative measurement of polarizabilities of the other alkali-metal atoms.

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**I. INTRODUCTION**

The electric dipole polarizability  $\alpha_E$  characterizes the response of the system to the homogeneous external electric field. It is related to various physical phenomena such as van der Waals interactions in ultracold collisions [1–3], Bose-Einstein condensation [4–7], binding positrons to atoms [8], and atomic energy shifts from the ambient blackbody radiation field in atomic clocks [9,10].

Polarizabilities for alkali-metal atoms can be determined experimentally with interferometric methods [11–15]. Recent advances in cooling techniques have allowed one to obtain the polarizability for the cesium ground state with 0.14% uncertainty [16]. However, such accuracy is not accessible at present for other alkali-metal systems. In contrast, the polarizability ratio for two different atoms can in some cases be measured more accurately than for a single atom [14,17,18]. This allows an improvement in determining polarizabilities from the ratio and the reference value. An appropriate candidate is the cesium atom [16]. However, the ratio measurement cannot be performed precisely with lighter atomic systems as yet. Another possibility is the accurate theoretical prediction for one of the light alkali-metal atoms, which can be set as the reference system. The most promising are calculations for the lithium atom, which can be done very accurately within nonrelativistic quantum electrodynamics (NRQED).

Nonrelativistic quantum electrodynamics is based on expansion of quantum electrodynamics in powers of the fine-structure constant  $\alpha$ . All the effects in NRQED expansion are expressed in terms of mean values with the wave function. Numerical solution of the Schrödinger equation can be obtained accurately for low-lying states of three-electron atom with explicitly correlated Hylleraas functions [19–22]. At the leading (nonrelativistic) order, the ground-state polarizability of the lithium atom has already been calculated with high numerical precision to be  $\alpha_0 = 164.112(1)$  [23]. The result  $\alpha_0 = 164.1117$  of comparable accuracy has also been obtained with explicitly correlated Gaussian (ECG) functions [24]. Calculations beyond the leading order have never been performed so far, probably due to high complexity. For relativistic

corrections only estimations exist, which lead to the total result  $\alpha_E = 164.11(3)$  [25]. Other treatments based on the relativistic coupled-cluster method [26] and on the relativistic many-body perturbation theory [27] do not include electron correlations accurately enough to compete with methods, which are based on the NRQED approach and explicitly correlated functions.

In this paper we provide the following benchmark for  $\alpha_0 = 164.112459(3)$  and also the reference value including the relativistic correction and the estimation of the leading QED effects  $\alpha_E = 164.0740(5)$ . The calculations are similar to those performed previously for the ground state of the helium atom [28], though significant modifications have to be introduced for the more complex system, such as the lithium atom. In order to improve the numerical precision of the relativistic corrections, we have derived a regularized representation for operators of the Breit-Pauli Hamiltonian in higher orders of perturbation theory. The accuracy of our result is limited only by the approximate treatment of the Bethe logarithm term [28], which has been verified in helium [29] to be well within the 10% uncertainty assumed here. The higher-order  $O(\alpha^6)$  QED corrections can safely be neglected. Apart from the leading QED corrections, we include also the leading finite nuclear mass corrections. As a result, our theoretical value for  $\alpha_E$  is two orders of magnitude more accurate than the best previous value in Ref. [25] and we recommend it as a reference for relative measurement of static polarizabilities.

All quantities given in this work are reported in atomic units.

**II. THEORY**

The nonrelativistic Hamiltonian for the lithium atom with an infinitely heavy nucleus and in the presence of an external electric field  $\mathcal{E}$  is given by

$$H = \sum_a \frac{p_a^2}{2} - \sum_a \frac{Z}{r_a} + \sum_{a>b} \frac{1}{r_{ab}} - \sum_a \mathcal{E}^i r_a^i, \quad (1)$$

where the summation convention is used for Cartesian coordinates. With this Hamiltonian, the nonrelativistic electric dipole

polarizability  $\alpha_0$  can be evaluated in second-order perturbation theory according to the formula

$$\alpha_0 = -\frac{2}{3} \sum_{a,b} \langle \phi_0 | r_a^i \frac{1}{E_0 - H_0} r_b^j | \phi_0 \rangle, \quad (2)$$

where  $\phi_0$  and  $E_0$  are the ground-state wave function and nonrelativistic energy, respectively, and  $H_0$  represents the Hamiltonian in Eq. (1) without the external field. The static polarizability beyond the nonrelativistic approximation can be obtained by modifying  $H_0$  by  $\delta H$ , which includes corrections due to the finite mass of the nucleus, relativity, and QED,

$$\delta H = \lambda H_{\text{MP}} + \alpha^2 H_{\text{rel}} + \alpha^3 H_{\text{QED}}, \quad (3)$$

where  $\lambda = -\mu/M$  is the ratio of the reduced electron mass to the nucleus mass and the subscript MP denotes the mass polarization correction. Using  $\delta H$  one finds a perturbative formula for the electric dipole polarizability

$$\alpha_E = \alpha_0 + \delta\alpha_E, \quad (4)$$

where

$$-\frac{3}{2} \delta\alpha_E = \sum_{a,b} \left[ 2 \langle \phi_0 | \delta H \frac{1}{(E_0 - H_0)} r_a^i \frac{1}{E_0 - H_0} r_b^j | \phi_0 \rangle + \langle \phi_0 | r_a^i \frac{1}{E_0 - H_0} (\delta H - \langle \delta H \rangle) \frac{1}{E_0 - H_0} r_b^j | \phi_0 \rangle \right]. \quad (5)$$

Components of  $\delta H$  are defined as (see, e.g., Ref. [30])

$$H_{\text{MP}} = -\sum_{a < b} p_a^i p_b^j, \quad (6)$$

$$H_{\text{rel}} = \sum_a \left[ -\frac{p_a^4}{8} + \frac{\pi Z}{2} \delta^3(r_a) \right] + \sum_{a < b} \left[ \pi \delta^3(r_{ab}) - \frac{1}{2} p_a^i \left( \frac{\delta^{ij}}{r_{ab}} + \frac{r_{ab}^i r_{ab}^j}{r_{ab}^3} \right) p_b^j \right]. \quad (7)$$

The QED correction is given by

$$H_{\text{QED}} = \frac{4Z}{3} \left[ \frac{19}{30} + \ln(\alpha^{-2}) - \ln k_0 \right] \sum_a \delta^3(r_a) + \left[ \frac{164}{15} + \frac{14}{3} \ln \alpha \right] \sum_{a < b} \delta^3(r_{ab}) - \frac{7}{6\pi} \sum_{a < b} P \left( \frac{1}{r_{ab}^3} \right), \quad (8)$$

where the Bethe logarithm  $\ln k_0$  has the form

$$\ln k_0 \equiv \frac{\langle \sum_a p_a^i (H_0 - E_0) \ln[2(H_0 - E_0)] \sum_b p_b^j \rangle}{2\pi Z \sum_c \langle \delta^3(r_c) \rangle} \quad (9)$$

and  $P(1/r_{ab}^3)$  is defined by

$$\langle \phi | P \left( \frac{1}{r^3} \right) | \psi \rangle = \lim_{a \rightarrow 0} \int d^3r \phi^*(\mathbf{r}) \left[ \frac{1}{r^3} \Theta(r - a) + 4\pi \delta^3(r) (\gamma + \ln a) \right] \psi(\mathbf{r}). \quad (10)$$

The corresponding terms in Eq. (5) can be systematically ordered as

$$\delta\alpha_E = \lambda A^{(0,1)} + \alpha^2 A^{(2,0)} + \alpha^3 A^{(3,0)} + \dots, \quad (11)$$

where  $A^{(m,n)}$  are the expansion coefficients of  $\alpha^m \lambda^n$ .

### III. DETAILS OF CALCULATIONS

The spatial part of the nonrelativistic ground-state wave function of lithium  $\phi_0$  is built as a linear combination of  $S$ -type Hylleraas functions

$$\phi = e^{-w_1 r_1 - w_2 r_2 - w_3 r_3} r_{23}^{n_1} r_{31}^{n_2} r_{12}^{n_3} r_1^{n_4} r_2^{n_5} r_3^{n_6}, \quad (12)$$

with non-negative integers  $n_i$  satisfying the condition

$$\sum_{i=1}^6 n_i \leq \Omega \quad (13)$$

for a maximum shell  $\Omega$  in the range from 7 up to 10. The whole basis set is divided into five sectors, each one with its own set of variational parameters  $w_i$  [20]. To avoid numerical instabilities, within each sector we drop the terms with  $n_4 > n_5$  (or  $n_4 < n_5$ ) and for  $n_4 = n_5$  drop terms with  $n_1 > n_2$  (or  $n_1 < n_2$ ). For each  $\Omega$ , the nonrelativistic energy was optimized with respect to the free parameters  $w_i$ . Details of the variational method with the Hylleraas basis can be found in our previous paper on the lithium ground state [21] and more accurate results are in Ref. [31]. Recently, Wang *et al.* published a work on large-scale Hylleraas calculations with modified sector decomposition with 26 520 basis elements generated with  $\Omega = 15$  [32]. Their result confirms the Hylleraas method as outstanding in applications for low-lying states of lithium atom with an uncertainty as small as  $5 \times 10^{-15}$ .

The first- and second-order perturbation correction to the wave function present in the formulas in Eqs. (2) and (5) can be denoted as

$$|\phi_1^i\rangle = \sum_a \frac{1}{E_0 - H_0} r_a^i |\phi_0\rangle, \quad (14)$$

$$|\phi_2\rangle = \sum_a \frac{1}{(E_0 - H_0)^2} r_a^i |\phi_1^i\rangle. \quad (15)$$

TABLE I. Nonrelativistic energies and electric dipole polarizabilities. Hyll. denotes the Hylleraas basis and ECG denotes explicitly correlated Gaussian calculations.

$\Omega$	$E_0$	$\alpha_0$
7	-7.478 060 311 577 67	164.112 426 9
8	-7.478 060 322 075 18	164.112 443 0
9	-7.478 060 323 864 52	164.112 458 3
10	-7.478 060 323 902 25	164.112 458 8
$\infty$		164.112 459(3)
ECG <sup>a</sup>	-7.478 060 323 81	
Hyll. <sup>b</sup>	-7.478 060 323 910 2(2)	
Hyll. <sup>c</sup>	-7.478 060 323 910 143 7(45)	
ECG <sup>d</sup>		164.111 7
Hyll. <sup>e</sup>		164.112(1)

<sup>a</sup>Reference [33].

<sup>b</sup>Reference [31].

<sup>c</sup>Reference [32].

<sup>d</sup>Reference [24].

<sup>e</sup>Reference [23].

TABLE II. Mean values  $\langle \phi_0 | A | \phi_2 \rangle$ . An asterisk denotes results for the Dirac  $\delta$  function obtained with a plain evaluation.

$\Omega$	$p_a^4$	$\delta(r_a)$	$\delta(r_{ab})$	$p_a^i r_{ab}^{-1} (\delta^{ij} + r_{ab}^i r_{ab}^j r_{ab}^{-2}) p_b^j$	$p_a^i p_b^i$	$r_{ab}^{-3}$
8	-8246.207	-153.250 37	-11.590 551 5	-19.945 53	-19.579 376	-82.534
9	-8246.438	-153.251 61	-11.590 426 8	-19.947 81	-19.579 378	-82.485
10	-8246.375	-153.249 66	-11.590 430 4	-19.948 13	-19.579 404	-82.445
11	-8246.278	-153.247 61	-11.590 444 9	-19.948 37	-19.579 346	-82.600
11		-153.241 24*	-11.587 700 3*			

The function in Eq. (14) is represented in terms of  $P$ -type Hylleraas basis functions as

$$\phi_a^i = r_a^i e^{-w_1 r_1 - w_2 r_2 - w_3 r_3} r_{23}^{n_1} r_{31}^{n_2} r_{12}^{n_3} r_1^{n_4} r_2^{n_5} r_3^{n_6}. \quad (16)$$

The first-order correction  $\phi_1^i$  is represented in two blocks of functions, which are relevant to core and external electrons, respectively. Each block contains functions up to the shell  $\Omega + 1$  and is divided into two sectors. Then the parameters  $w_i$  are optimized to achieve the best upper-bound value of  $\alpha_0$ . For second-order correction  $\phi_2$  we use  $S$ -type functions limited by the  $(\Omega + 1)$  shell, which are divided into five sectors with the same prescription as in the construction of the wave function  $\phi_0$ .

Numerical results are presented in Table I. Our value for  $\alpha_0$  is almost three orders of magnitude more accurate than previously published in Ref. [23]. A possible explanation for such an improvement is the careful optimization of the nonlinear parameters  $w_i$ . With accurate result for  $\alpha_0$  and the first-order correction to the wave function  $\phi_1^i$  we are able to include small effects, thus refining theoretical predictions of  $\alpha_E$ . The objective of the calculations are finite-mass, leading relativistic, and QED corrections to  $\delta\alpha_E$  using Eq. (5), which can be rewritten in the form

$$-\frac{3}{2}\delta\alpha_E = 2\langle \phi_0 | \delta H | \phi_2 \rangle + \langle \phi_1^i | (\delta H - \langle \delta H \rangle) | \phi_1^i \rangle. \quad (17)$$

It involves the evaluation of different mean values. We presented details of the first-order calculations  $\langle \phi_0 | \delta H | \phi_0 \rangle \equiv \langle \delta H \rangle$  in Ref. [22] and the most accurate results have been published in Ref. [34]. The only exception is the value of the Bethe logarithm defined in Eq. (9), which was published by Yan *et al.* [35].

Direct use of Eqs. (8) and (9) leads to a slow numerical convergence. This is observed for operators sensitive to short-distance inaccuracies of the wave function such as  $p_a^4$  and the Dirac  $\delta$  function. The solution is to regularize matrix elements

of these operators, following Drachman's recipes [36], in the higher-order perturbation formulas. In the paper we provide the set of formulas for the leading relativistic correction to  $\alpha_0$ . The following operators have been used in the regularization of the Breit-Pauli Hamiltonian [22]:

$$\sum_a [p_a^4]_r = 4(E_0 - V)^2 - \sum_{a \neq b} p_a^2 p_b^2, \quad (18)$$

$$4\pi \sum_a [\delta(r_a)]_r = 4 \sum_a (E_0 - V) r_a^{-1} - 2 \sum_{a,b} p_b^i r_a^{-1} p_b^i, \quad (19)$$

$$4\pi \sum_{a < b} [\delta(r_{ab})]_r = 2 \sum_{a < b} (E_0 - V) r_{ab}^{-1} - \sum_{a < b, c} p_c^i r_{ab}^{-1} p_c^i. \quad (20)$$

We present formulas for the components of  $\delta\alpha_E$  in Eq. (5),

$$\sum_a \langle \phi_0 | p_a^4 | \phi_2 \rangle = \sum_a \langle \phi_0 | [p_a^4]_r | \phi_2 \rangle - 4 \sum_b \langle \phi_0 | (2E_0 - V) r_b^i | \phi_1^i \rangle, \quad (21)$$

$$\sum_a \langle \phi_1^i | p_a^4 | \delta\phi_1^i \rangle = \sum_a \langle \phi_1^i | [p_a^4]_r | \phi_1^i \rangle - 8 \sum_b \langle \phi_0 | (E_0 - V) r_b^i | \phi_1^i \rangle + 12 \sum_a \langle r_a^2 \rangle - 4 \sum_{a < b} \langle r_{ab}^2 \rangle, \quad (22)$$

$$\sum_a \langle \phi_0 | 4\pi \delta(r_a) | \phi_2 \rangle = \sum_a \langle \phi_0 | 4\pi [\delta(r_a)]_r | \phi_2 \rangle - 2 \sum_{a,b} \langle \phi_0 | (r_a^{-1} - r_b^{-1}) r_b^i | \phi_1^i \rangle, \quad (23)$$

TABLE III. Mean values  $\langle \phi_1^i | A - \langle A \rangle | \phi_1^i \rangle$ . An asterisk denotes results for the Dirac  $\delta$  function obtained with a plain evaluation.

$\Omega$	$p_a^4$	$\delta(r_a)$	$\delta(r_{ab})$	$p_a^i r_{ab}^{-1} (\delta^{ij} + r_{ab}^i r_{ab}^j r_{ab}^{-2}) p_b^j$	$p_a^i p_b^i$	$r_{ab}^{-3}$
8	-28 732.39	-561.641 21	-41.097 594	-224.650 40	-152.105 776 9	-19.63
9	-28 732.70	-561.635 21	-41.097 513	-224.659 71	-152.105 735 8	-18.30
10	-28 733.04	-561.634 93	-41.097 454	-224.662 60	-152.105 738 3	-17.60
11	-28 733.21	-561.635 57	-41.097 423	-224.663 45	-152.105 725 8	-17.34
11		-561.643 85*	-41.090 327*			

TABLE IV. Final value, numerical coefficients, and contributions related to the fine-structure constant  $\alpha$  expansion of the electric dipole polarizability  $\alpha_E$  for  ${}^7\text{Li}$ . The mass of the nucleus  $M({}^7\text{Li}) = 7.016\,003\,425\,6(45)$  [37]. A comparison to theoretical and experimental data is presented. Except for the  $\alpha^3$  coefficient  $A^{(3,0)}$ , other uncertainties are of numerical origin.

Order	$A^{(m,n)}$	Contribution
1	164.112 459(3)	164.112 46
$\alpha^2$	-995.06(2)	-0.052 99
$\alpha^3$	11 700.(1200)	0.004 5(5)
$\lambda$	-127.509 64(12)	0.009 97
Total		164.074 0(5)
Theor. <sup>a</sup>		163.74
Theor. <sup>b</sup>		164.08
Theor. <sup>c</sup>		164.11(3)
Expt. <sup>d</sup>		164.0(3.4)
Expt. <sup>e</sup>		164.19(1.08)

<sup>a</sup>References [26].

<sup>b</sup>References [27].

<sup>c</sup>References [35].

<sup>d</sup>References [12].

<sup>e</sup>References [11,38].

$$\sum_a \langle \phi_1^i | 4\pi \delta(r_a) | \phi_1^i \rangle = \sum_a \langle \phi_1^i | 4\pi [\delta(r_a)]_r | \phi_1^i \rangle - 4 \sum_{a,b} \langle \phi_0 | r_a^{-1} r_b^i | \phi_1^i \rangle, \quad (24)$$

$$\sum_{a<b} \langle \phi_0 | 4\pi \delta(r_{ab}) | \phi_2 \rangle = \sum_{a<b} \langle \phi_0 | 4\pi [\delta(r_{ab})]_r | \phi_2 \rangle - \sum_{a<b,c} \langle \phi_0 | (r_{ab}^{-1} - (r_{ab}^{-1})) r_c^i | \phi_1^i \rangle, \quad (25)$$

$$\sum_{a<b} \langle \phi_1^i | 4\pi \delta(r_{ab}) | \phi_1^i \rangle = \sum_{a<b} \langle \phi_1^i | 4\pi [\delta(r_{ab})]_r | \phi_1^i \rangle - 2 \sum_{a<b,c} \langle \phi_0 | r_{ab}^{-1} r_c^i | \phi_1^i \rangle, \quad (26)$$

and numerical results are collected in Tables II and III. For Dirac- $\delta$  operators we also present nonregularized results. The accuracy of such mean values is at least one order of magnitude lower than regularized expressions. The enhancement seems to be even more remarkable for two electron operators. Although the calculations with regularized forms are much more complicated, the effort is justified in the case of lithium. It significantly reduces the uncertainty of the total result presented in Table IV. This is different in comparison to the helium atom, where regularized methods are not mandatory

due to much higher numerical precision of the wave function [28,29].

#### IV. FINAL RESULTS

The obtained  $\alpha_E$  improves the accuracy of theoretical predictions [25] since it takes into account the leading relativistic and QED effects. It is in accordance with the result calculated within the relativistic many-body theory [27], which is less precise due to inexact treatment of electron correlations. Moreover, it is also in good agreement with experimental values [11,12,38], although at present they are much less accurate. The uncertainty of our result comes from the approximate treatment of the correction related to the Bethe logarithm. To estimate this effect, we use the value for a free atom  $\ln k_0(2^1S) = 5.178\,28(1)$  [35], thus neglecting the dependence of the Bethe logarithm on the electric field, and assume an uncertainty of 10%, which corresponds to a  $5 \times 10^{-4}$  absolute uncertainty of the final result. The higher-order QED corrections are at present negligible. The dominating part, known from the one-loop self-energy in hydrogenic systems

$$H_{\text{HQED}} = Z^2 \left[ \frac{427}{96} - 2 \ln(2) \right] \sum_a \delta^3(r_a), \quad (27)$$

gives about a  $6.3 \times 10^{-5}$  correction to the polarizability, which is much smaller than the uncertainty of the Bethe logarithmic contribution.

#### V. CONCLUSION

We have calculated the electric dipole polarizability  $\alpha_E$  of the lithium ground state, on the basis of the NRQED approach, using explicitly correlated Hylleraas functions. Our value, the most accurate to date, is in agreement with previous ones and can be considered as a benchmark for the relativistic coupled-cluster and relativistic many-body perturbation theory methods. The experimental accuracy is not yet good enough to verify our theoretical predictions, with one exception. The relative measurement of polarizabilities between two states of the same atom, such as for  ${}^7\text{Li}$ ,  $\alpha_E(2^1S) - \alpha_E(2^1P) = 37.146(17)$  by Hunter *et al.* [39], can reach competitive accuracy. For the comparison with this experimental value, the polarizability  $\alpha_E$  of the  $2^1P$  state has to be calculated including relativistic and QED corrections, and this has not yet been done.

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