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Ab initio determination of the polarizability of neon

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The static electric-dipole polarizability α of the neon atom was determined with a relative uncertainty of only about 0.003% using state-of-the-art *ab initio* approaches. The new value, $\alpha = 2.661\,067(77)$ a.u., is almost five times more accurate than the previous *ab initio* estimate, $\alpha = 2.660\,80(36)$ a.u., by Lesiuk *et al.* [Phys. Rev. A **102**, 052816 (2020)]. Similar to their work, we calculated α using *ab initio* methods up to full configuration interaction and added corrections for finite nuclear mass and size, relativistic, and quantum electrodynamics (QED) effects. The uncertainty reduction of this work was achieved in particular by employing extremely large basis sets, including newly developed ones of 11Z, 12Z, and 13Z quality. Moreover, the finite nuclear mass effects and most of the relativistic contributions were calculated at much higher levels of theory than in the work of Lesiuk *et al.* However, we adopted their values for the orbit-orbit part of the relativistic correction and for the Bethe logarithm needed to compute the QED correction. The uncertainty of our final value is still an order of magnitude larger than that of the experimental value recently measured by Gaiser and Fellmuth [Phys. Rev. Lett. **120**, 123203 (2018)] with an uncertainty of only a few parts per million using dielectric-constant gas thermometry. Yet, our *ab initio* value agrees with their value, $\alpha_{\text{exp}} = 2.661\,057(7)$ a.u., almost within the experimental uncertainty. This could indicate that the higher-order relativistic corrections and QED contributions, which dominate our uncertainty budget, are more accurate than expected considering the uncontrolled approximations involved in their calculation.

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I. INTRODUCTION

The static electric-dipole polarizability α is one of the most important properties of an atom or a molecule that governs its interaction with an external electric field. For noble gases, such as neon, α is a scalar quantity, which can be defined by

$$\alpha = \lim_{\mathscr{E}\to 0}\left(\frac{\mu_{\text{ind}}}{\mathscr{E}}\right), \tag{1}$$

where μ_{ind} is the strength of the atomic dipole moment induced by a static, homogeneous external electric field of strength $\mathscr{E}$. The polarizability of a noble gas is directly related to its relative permittivity ε_{r} in the dilute gas limit through the well-known Clausius-Mossotti relation, which in SI units is given as

$$\frac{\varepsilon_{\text{r}} - 1}{\varepsilon_{\text{r}} + 2} = \frac{N\alpha}{3\varepsilon_0}, \tag{2}$$

where N is the number density of the gas and ε_0 is the vacuum permittivity. If N is expressed through the ideal gas law, one obtains a relation between the relative permittivity ε_{r}, the thermodynamic temperature T, and the pressure p. Provided that α is accurately known, the thermodynamic temperature of the gas can be determined by measuring ε_{r} (through the capacitance of a capacitor filled with the gas) as a function of p along an isotherm, followed by extrapolation to zero pressure. Measuring thermodynamic temperature in this manner is referred to as dielectric-constant gas thermometry (DCGT) [1]. Similarly, if ε_{r} and T are measured and α is known, p can be determined [2], or if ε_{r}, T, and p are measured, α can be determined [3,4]. The DCGT method is closely related to refractive-index gas thermometry [5,6], in which the refractive index instead of the relative permittivity is measured. The refractive index in the dilute gas limit is a function of the static electric-dipole polarizability, the magnetic susceptibility, and in some cases (depending on the wavelength at which the measurements are performed) the frequency dependence of the electric-dipole polarizability.

Recently, Gaiser and Fellmuth [4] used DCGT to measure the static electric-dipole polarizabilities of helium, neon, and argon with unprecedented low standard uncertainties of only about 2 ppm. Helium has been the noble gas of choice so far in DCGT because its polarizability is known extremely accurately from theory [7–9], and the value obtained from the new measurements, while not as accurate as the theoretical one [9], is fully consistent with it. The primary motivation of Gaiser and Fellmuth [4] for including also neon and argon was to enable these two gases to be used as alternatives to helium in standard DCGT applications. The use of helium has the disadvantage that its polarizability is extremely small, which leads to a strong sensitivity to impurities. This sensitivity can be reduced by using neon, which has approximately twice the polarizability of helium, or argon, which is about eight times more polarizable than helium. A further motivation for the work of Gaiser and Fellmuth [4] was to give theoreticians accurate reference polarizabilities to allow them to stringently

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test their *ab initio* methodologies (and if necessary improve them) for neon and argon. Existing theoretical polarizability values for these two gases were multiple orders of magnitude less accurate than the new measured values.

Shortly after the work of Gaiser and Fellmuth was published, Lesiuk *et al.* [10] presented a new *ab initio* polarizability value for neon, which is much more accurate than any previous theoretical value because for the first time all relevant physical effects were systematically considered. Their starting point was a nonrelativistic calculation of the polarizability using the orbital-unrelaxed linear-response CC3 method [11], which is a variant of coupled-cluster (CC) theory. The calculations were performed with purpose-built Slater-type orbital basis sets up to 7Z quality, and the results were extrapolated to the complete basis set (CBS) limit. They then calculated corrections for higher coupled-cluster levels up to the full configuration interaction (FCI) limit with smaller Gaussian-type orbital basis sets. Moreover, they added corrections for finite nuclear mass and size effects as well as the relativistic contributions of order $1/c^2$ (where $c = 137.036$ is the speed of light in atomic units) and quantum electrodynamics (QED) corrections of order $1/c^3$ and $1/c^4$. However, their uncertainty estimate for the final polarizability value is still more than 50 times larger than that given by Gaiser and Fellmuth [4] for their experimental value, but the agreement between the two polarizability values is within the uncertainty estimated by Lesiuk *et al.* [10].

In this work, we present a new *ab initio* value for the polarizability of neon with significantly reduced uncertainty compared with the result of Lesiuk *et al.* [10]. For the calculations, we employed standard quantum chemistry codes, although for a few polarizability contributions we had to adopt results calculated by Lesiuk *et al.* [10] using specialized in-house codes, namely, for the orbit-orbit term in the relativistic contribution of order $1/c^2$ and for the Bethe logarithm needed to compute the QED correction of order $1/c^3$.

The aimed uncertainty reduction required the use of extremely large basis sets. For neon, Gaussian-type orbital basis sets up to 10Z quality are available in the literature, but we developed even larger ones up to 13Z quality and modified and/or reoptimized some of the existing basis sets. In Sec. II, we therefore first discuss the basis sets used in this work before presenting the actual polarizability calculations in Sec. III. In Sec. IV, we discuss the results in comparison with those of Lesiuk *et al.* [10] and of Gaiser and Fellmuth [4] and draw conclusions.

Note that we use atomic units (a.u.) throughout the remainder of this paper.

II. BASIS SETS

We used two different families of Gaussian basis sets for the calculations reported in this work. For contributions for which only relatively small basis sets were affordable, we employed the standard correlation consistent basis sets cc-pVnZ with $2 \leqslant n \leqslant 6$ supplemented by either one layer of diffuse functions (aug-cc-pVnZ), two layers of diffuse functions (d-aug-cc-pVnZ), or three layers of diffuse functions (t-aug-cc-pVnZ) [12–15]. For calculations in which all electrons were correlated, the (d-, t-)aug-cc-pCVnZ basis sets [16,17], which contain additional tight basis functions, were used in uncontracted form and without the additional tight s and p primitive functions to avoid near-linear-dependency issues (the uncontraction of the basis sets yields already plenty of tight primitive s and p functions). These basis sets are denoted as (d-, t-)aug-u-cc-pCVnZ.

For calculations for which it was possible to employ larger basis sets, we initially considered using again the cc-pVnZ basis sets, which are available for neon up to $n = 10$ [18–20]. However, there are apparent irregularities in the series for high n values. As an example, the largest primitive s exponent, which should increase in a smooth fashion with increasing n, has values of 7.149 for cc-pV6Z [14], 10.8323 for cc-pV7Z [18], and 17.0553 for cc-pV8Z [19], but it then remains almost at the same value, 17.1191, for cc-pV9Z [20]. A similar behavior can be observed for the p exponents. The reason for these irregularities is that the cc-pVnZ basis sets generate the s and p primitives by splitting off functions from the contracted s and p sets optimized at the Hartree-Fock self-consistent-field (SCF) level of theory, but for cc-pV9Z the exponents of the primitives in the contracted sets are not properly optimized. When we performed initial test calculations of the polarizability of neon at a correlated level of theory with these basis sets (supplemented by suitable diffuse functions), we observed that the resulting polarizability values also behaved in a somewhat irregular manner when going from the 8Z to the 9Z basis set level. Thus, with these basis sets, it is not possible to extrapolate properly to the CBS limit, which is necessary for several polarizability contributions. Therefore, we did not further consider the cc-pVnZ basis sets for $n > 6$.

A more recently developed alternative to the cc-pVnZ basis sets is the nZaP family of basis sets [21–23], which is available for neon also up to $n = 10$ [21]. The major distinction between the two families is that the nZaP basis sets have, in addition to the contracted SCF-optimized s and p basis functions, separate even-tempered sets of s and p primitives optimized by minimizing the total energy obtained using second-order Møller-Plesset perturbation theory (MP2) in the frozen-core (FC) approximation. The higher angular-momentum functions also form even-tempered sets optimized at the FC-MP2 level of theory, whereas the cc-pVnZ basis sets use even-tempered higher angular-momentum sets optimized at the FC configuration interaction with single and double excitations level of theory. The nZaP basis sets have one diffuse primitive s function and one set of diffuse primitive p functions (the "a" in nZaP), generated for neon from the lowest two exponents in each of the contracted s and p basis functions in an even-tempered manner. The respective basis sets that also have diffuse functions of higher angular momentum l are called nZaPa [23], where the additional exponents up to the second-highest angular momentum $l = n - 1$ were generated from the existing two smallest exponents of each angular momentum again in an even-tempered manner, while no diffuse functions were added for $l = n$ [23].

In the present work, we made some modifications to the existing nZaP and nZaPa basis sets for neon. First, because we noticed small but not insignificant irregularities in the exponents also for this family of basis sets, we tried to further optimize the exponents of the primitive functions (while still

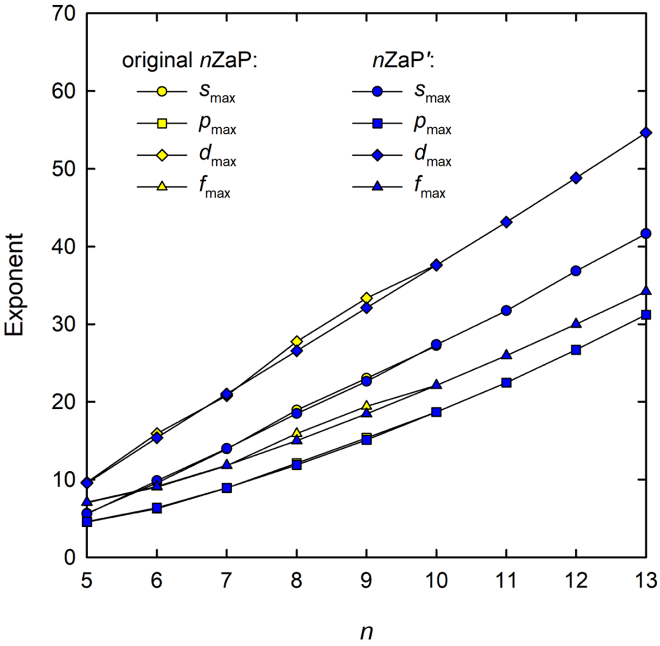

FIG. 1. Highest s, p, d, and f exponents of the primitive basis functions in the original nZaP basis sets [21] for $5 \leqslant n \leqslant 10$ and in the reoptimized ($5 \leqslant n \leqslant 10$) and new ($11 \leqslant n \leqslant 13$) basis sets of this work, nZaP′. The connecting lines are shown for clarity.

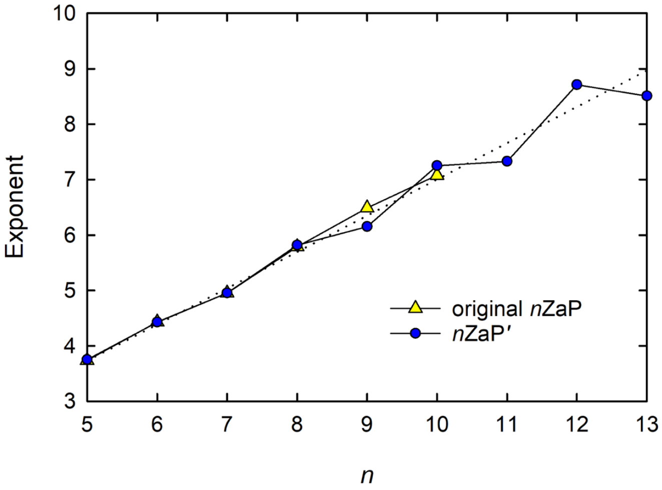

FIG. 2. Exponent of the set of functions with the highest angular momentum $l = n$ in the original nZaP basis sets [21] for $5 \leqslant n \leqslant 10$ (e.g., for $n = 5$ the exponent of the single set of h functions) and in the reoptimized ($5 \leqslant n \leqslant 10$) and new ($11 \leqslant n \leqslant 13$) basis sets of this work, nZaP′. The connecting lines and the dotted line are shown for clarity.

keeping them even tempered) for the 5ZaP to 10ZaP basis sets. The 9ZaP basis set [21] was published with a wrong exponent for the single diffuse s function (it had the same exponent as in the 8ZaP basis set), which we corrected before reoptimizing the basis set. For 5ZaP, the achieved FC-MP2 energy lowering is only about $1.5 \times 10^{-7}\,E_\mathrm{h}$, but for the higher basis sets it is more significant. The largest lowering of approximately $2.0 \times 10^{-5}\,E_\mathrm{h}$ was achieved for 10ZaP, for which the most drastic change in any of the individual exponents was found for the higher of the two m function exponents, decreasing from 11.9658 to 8.7947. The reoptimized basis sets, which we believe to be optimized to within $1 \times 10^{-9}\,E_\mathrm{h}$, are denoted as nZaP′. In Fig. 1, the changes in the exponents are exemplarily shown for the highest exponents of the primitive functions of s, p, d, and f symmetry. The figure demonstrates that the n dependence is distinctly smoother after the reoptimization. In the next step, we removed the diffuse s and p functions from the basis sets and added new diffuse functions, which we generated by multiplying the smallest exponent for each angular momentum (including the highest) by a factor of 0.5. The resulting basis sets are denoted as aug-nZP′ (without diffuse functions, they are denoted simply as nZP′). Doubly augmented versions of these basis sets, d-aug-nZP′, were generated from the augmented ones in the same manner, and we continued this procedure until reaching quadruply augmented basis sets, q-aug-nZP′.

In addition to modifying already existing basis sets, we also developed nZP′ basis sets for $n = 11$ to $n = 13$. In the first step, the exponents of the primitive sets ($27s20p$), ($29s22p$), and ($31s24p$), intended for $n = 11$, 12, and 13, respectively, were fully optimized by minimizing the SCF energy and then contracted to [$2s1p$]. The size of each set of primitives was chosen such that the increment pattern established by the existing 9ZaP and 10ZaP basis sets is continued. The smallest s and p exponents in each of the new sets were then used for generating a single diffuse s function and a single set of diffuse p functions to adhere to the scheme used for constructing the original nZaP basis sets [21]. Following again the original scheme [21], the required even-tempered sets of primitive functions for all angular momenta were added and optimized by minimizing the FC-MP2 energy, resulting in 11ZaP′, 12ZaP′, and 13ZaP′ basis sets. The highest exponents of the primitive functions of s, p, d, and f symmetry for the three basis sets are included in Fig. 1 to illustrate the smooth behavior of these exponents for the full range of n values up to $n = 13$. In the final step, the diffuse s and p functions were removed again and replaced by new diffuse functions generated as described above. Already the 13ZP′ basis set, which has no dedicated diffuse functions, is certainly one of the largest Gaussian basis sets developed so far. In addition to the [$2s1p$] set, it has 12 s, 12 p, 12 d, 11 f, 10 g, 9 h, 8 i, 7 k, 6 l, 5 m, 4 n, 3 o, 2 q, and 1 r set of primitive functions. In the case of the q-aug-13ZP′ basis set, each of these numbers of primitive sets is increased by 4.

In Fig. 2, we show an interesting pattern in the values for the exponent of the set of functions with the highest angular momentum in the nZaP′ basis sets, where the values for even n are always above and for odd n always below a straight trend line. This results, for example, in the exponent of the single set of r functions in 13ZaP′ being actually smaller than the exponent of the single set of q functions in 12ZaP′, despite the trend line rising. This behavior is not apparent for the original nZaP basis sets.

For calculations with all electrons correlated, we removed the even-tempered s and p primitives from the nZP′ basis sets and uncontracted the basis sets. For each angular momentum l from $l = 2$ (i.e., d symmetry) up to the third-highest angular momentum $l = n - 2$, two tight functions were generated by multiplying the highest exponent of the existing even-tempered sets by factors of 2 and 6. For the second-highest angular momentum $l = n - 1$, a single set of tight functions was generated by multiplying the higher exponent of the two existing sets of functions of this angular momentum by a factor of 3. For the highest angular momentum $l = n$, no tight functions were added. The first layer of diffuse functions was generated as described above by multiplying the smallest exponent for each angular momentum (including $l = 0$ and $l = 1$) by a factor of 0.5. Doubly and triply augmented sets were generated by repeating this procedure two times. The resulting basis sets are denoted as (d-, t-)aug-C*n*ZP′.

The SCF and MP2 calculations needed to reoptimize the existing basis sets and to develop the new ones were carried out with the PSI3 code [24]. Some of the correlation consistent basis sets for neon used in this work were obtained from the Basis Set Exchange [25–27]. The *n*ZaP basis sets up to $n = 10$ were taken from the supplement of the original publication [21]. The *n*ZP′ basis sets for $n = 5$ to $n = 13$ developed in this work are provided in the Supplemental Material [28].

III. CALCULATION OF THE POLARIZABILITY

A. General approach

The energy E of a noble gas atom in a static, homogeneous electric field with strength $\mathscr{E}$ can be written as

$$E(\mathscr{E}) = E_0 + \frac{1}{2}\left(\frac{\partial^2 E}{\partial \mathscr{E}^2}\right)_{\mathscr{E}=0} \mathscr{E}^2 + O(\mathscr{E}^4), \tag{3}$$

where E_0 is the energy of the atom in the absence of a field. With $\alpha = -(\partial^2 E/\partial \mathscr{E}^2)_{\mathscr{E}=0}$, we obtain

$$E(\mathscr{E}) = E_0 - \tfrac{1}{2}\alpha \mathscr{E}^2 + O(\mathscr{E}^4), \tag{4}$$

and hence

$$\frac{2[E_0 - E(\mathscr{E})]}{\mathscr{E}^2} = \alpha + O(\mathscr{E}^2). \tag{5}$$

Equation (5) allows the evaluation of α by calculating E_0 as well as E for at least two different small field strengths, followed by extrapolation of $2[E_0 - E(\mathscr{E})]/\mathscr{E}^2$ linearly in $\mathscr{E}^2$ to $\mathscr{E} = 0$. In this work, we applied field strengths of 0.005 and 0.01 a.u.

We note that the quantum-chemical program package CFOUR [29,30] can evaluate the second derivative $\alpha = -(\partial^2 E/\partial \mathscr{E}^2)_{\mathscr{E}=0}$ analytically for several high-level *ab initio* methods and thus allows a direct evaluation of α from a single calculation. Both orbital-relaxed and orbital-unrelaxed analytical derivative calculations are possible. The numerical second-derivative procedure with finite electric fields conforms to the orbital-relaxed analytical approach. Because the computational effort for the analytical implementation is much higher than for the numerical procedure and because the public version of CFOUR can only handle basis functions up to *i* symmetry, we used this analytical implementation only for a few test calculations to determine the accuracy of the numerical derivative approach. We found that the polarizabilities obtained with the two approaches agree to within about six to seven digits, which is sufficient for our purposes.

The total static electric-dipole polarizability is formulated as

$$\alpha = \alpha_{\mathrm{FCI}} + \Delta\alpha_{\mathrm{FNM}} + \Delta\alpha_{\mathrm{FNS}} + \Delta\alpha_{\mathrm{rel}} + \Delta\alpha_{\mathrm{QED}}, \tag{6}$$

where α_{FCI} is the nonrelativistic polarizability in the FCI limit assuming a point nucleus of infinite mass, $\Delta\alpha_{\mathrm{FNM}}$ and $\Delta\alpha_{\mathrm{FNS}}$ are the corrections for finite nuclear mass and size effects, and $\Delta\alpha_{\mathrm{rel}}$ and $\Delta\alpha_{\mathrm{QED}}$ are the corrections for relativistic and QED effects. Each of these terms is made up of several individual contributions, with the starting point being the polarizability at the SCF level of theory as part of α_{FCI}. Many of these individual contributions required extrapolation to the CBS limit. In most cases, we applied the standard two-parameter CBS extrapolation scheme recommended by Halkier *et al.* [31],

$$\Delta\alpha(n) = \Delta\alpha(\mathrm{CBS}) + \frac{a}{n^3}. \tag{7}$$

As is commonly done with this scheme, we only used the two highest basis set levels in the extrapolation. For some contributions, a third fit parameter was introduced,

$$\Delta\alpha(n) = \Delta\alpha(\mathrm{CBS}) + \frac{a}{n^3} + \frac{b}{n^4}, \tag{8}$$

and more than the minimally required three basis set levels were included in the fit with this formula to obtain more stable results. We tested a few alternative three-parameter schemes, but none were as simple and stable as the one given by Eq. (8).

In the following subsections, we discuss the calculation of the terms α_{FCI}, $\Delta\alpha_{\mathrm{FNM}}$, $\Delta\alpha_{\mathrm{FNS}}$, $\Delta\alpha_{\mathrm{rel}}$, and $\Delta\alpha_{\mathrm{QED}}$.

B. Nonrelativistic polarizability in the FCI limit assuming a point nucleus of infinite mass

The nonrelativistic polarizability in the FCI limit of a neon atom with a pointlike nucleus of infinite mass, α_{FCI}, is expressed here as a sum of two main contributions:

$$\alpha_{\mathrm{FCI}} = \alpha_{\mathrm{CCSD(T)}} + \Delta\alpha_{\mathrm{FCI-CCSD(T)}}, \tag{9}$$

where $\alpha_{\mathrm{CCSD(T)}}$ denotes the value for the coupled-cluster level with single, double, and perturbative triple excitations [CCSD(T)] [32], and $\Delta\alpha_{\mathrm{FCI-CCSD(T)}}$ is the correction for higher coupled-cluster contributions up to the FCI limit.

The contribution $\alpha_{\mathrm{CCSD(T)}}$ was split into five parts:

$$\begin{aligned}\alpha_{\mathrm{CCSD(T)}} = {}& \alpha_{\mathrm{SCF}} + \Delta\alpha^{\mathrm{FC}}_{\mathrm{OS\text{-}MP2}} + \Delta\alpha^{\mathrm{FC}}_{\mathrm{SS\text{-}MP2}} \\ & + \Delta\alpha^{\mathrm{FC}}_{\mathrm{CCSD(T)-MP2}} + \Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{CCSD(T)}},\end{aligned} \tag{10}$$

where α_{SCF} is the polarizability at the SCF level of theory, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{OS\text{-}MP2}}$ and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{SS\text{-}MP2}}$ denote the FC-MP2 correlation contributions from the opposite-spin (OS) and same-spin (SS) electron pairs, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{CCSD(T)-MP2}}$ is the polarizability difference between the FC-CCSD(T) and FC-MP2 levels of theory, and $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{CCSD(T)}}$ denotes the polarizability difference between the all-electron (AE) CCSD(T) and FC-CCSD(T) levels.

The contributions α_{SCF}, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{OS\text{-}MP2}}$, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{SS\text{-}MP2}}$, and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{CCSD(T)-MP2}}$ were obtained for the basis sets aug-*n*ZP′, d-aug-*n*ZP′, t-aug-*n*ZP′, and q-aug-*n*ZP′ with $n = 5$ to $n = 12$ from FC-CCSD(T) calculations. For $n = 13$, we only performed FC-MP2 calculations and, thus, did not obtain $\Delta\alpha^{\mathrm{FC}}_{\mathrm{CCSD(T)-MP2}}$. The results (in a.u.) are listed in Table I.

TABLE I. Calculated and extrapolated values for the polarizability at the SCF level of theory, α_{SCF}; for the OS and SS parts to the FC-MP2 correlation polarizability contribution, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{OS\text{-}MP2}}$ and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{SS\text{-}MP2}}$; and for the polarizability difference between the FC-CCSD(T) and FC-MP2 levels of theory, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{CCSD(T)-MP2}}$. In this table and the following tables, all values are in a.u., and the final values are given in bold, with the numbers in parentheses indicating the estimated uncertainties.

n	aug-nZP′	d-aug-nZP′	t-aug-nZP′	q-aug-nZP′
		α_{SCF}		
5	2.262392	2.374824	2.376657	2.376664
6	2.301248	2.376012	2.376729	2.376729
7	2.336013	2.376565	2.376744	2.376743
8	2.352704	2.376693	2.376745	2.376745
9	2.362290	2.376727	2.376744	2.376744
10	2.367146	2.376737	2.376744	2.376744
11	2.370282	2.376741	2.376744	2.376744
12	2.371939	2.376743	2.376744	2.376744
13	2.373176	2.376744	2.376745	**2.376745(1)**
		$\Delta\alpha^{\mathrm{FC}}_{\mathrm{OS\text{-}MP2}}$		
5	0.179505	0.219402	0.221096	0.221100
6	0.189780	0.218441	0.219204	0.219208
7	0.200338	0.217832	0.218118	0.218122
8	0.205976	0.217310	0.217450	0.217455
9	0.209537	0.216947	0.217025	0.217030
10	0.211461	0.216682	0.216736	0.216741
11	0.212789	0.216505	0.216542	0.216546
12	0.213513	0.216371	0.216400	0.216403
13	0.214084	0.216279	0.216300	0.216302
∞				**0.215897(8)**
		$\Delta\alpha^{\mathrm{FC}}_{\mathrm{SS\text{-}MP2}}$		
5	0.081299	0.103285	0.104152	0.104156
6	0.087572	0.103678	0.104061	0.104062
7	0.093918	0.103903	0.104021	0.104021
8	0.097421	0.103960	0.104001	0.104001
9	0.099678	0.103975	0.103990	0.103990
10	0.100936	0.103978	0.103984	0.103985
11	0.101813	0.103979	0.103981	0.103981
12	0.102307	0.103978	0.103979	0.103979
13	0.102695	0.103978	0.103978	0.103978
∞				**0.103976(1)**
		$\Delta\alpha^{\mathrm{FC}}_{\mathrm{CCSD(T)-MP2}}$		
5	−0.024106	−0.031554	−0.031790	−0.031802
6	−0.026847	−0.032419	−0.032560	−0.032566
7	−0.029388	−0.032805	−0.032881	−0.032887
8	−0.030798	−0.032984	−0.033037	−0.033042
9	−0.031684	−0.033080	−0.033120	−0.033123
10	−0.032182	−0.033136	−0.033167	−0.033171
11	−0.032514	−0.033173	−0.033198	−0.033200
12	−0.032708	−0.033197	−0.033218	−0.033218
∞				**−0.033265(3)**

Our best estimate for α_{SCF} is the value obtained with the q-aug-13ZP′ basis set,

$$\alpha_{\mathrm{SCF}} = 2.376\,745(1),$$

where the number in parentheses indicates our estimate of the uncertainty in the last digit. The other three contributions were extrapolated to the CBS limit. For $\Delta\alpha^{\mathrm{FC}}_{\mathrm{OS\text{-}MP2}}$ and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{CCSD(T)-MP2}}$, the extrapolation was performed by fitting Eq. (8) to the values for the q-aug-7ZP′ to q-aug-13ZP′ and q-aug-8ZP′ to q-aug-12ZP′ basis sets, respectively. The contribution $\Delta\alpha^{\mathrm{FC}}_{\mathrm{SS\text{-}MP2}}$ converges very quickly to the CBS limit, since in this case the error is not approximately proportional to n^{-3} but to n^{-5}. The extrapolation was performed using Eq. (7), but, consequently, with the exponent of n in the denominator changed from 3 to 5. Only the $\Delta\alpha^{\mathrm{FC}}_{\mathrm{SS\text{-}MP2}}$ values for $n = 12$ and $n = 13$ were used for this extrapolation. The resulting CBS estimates for the three contributions are

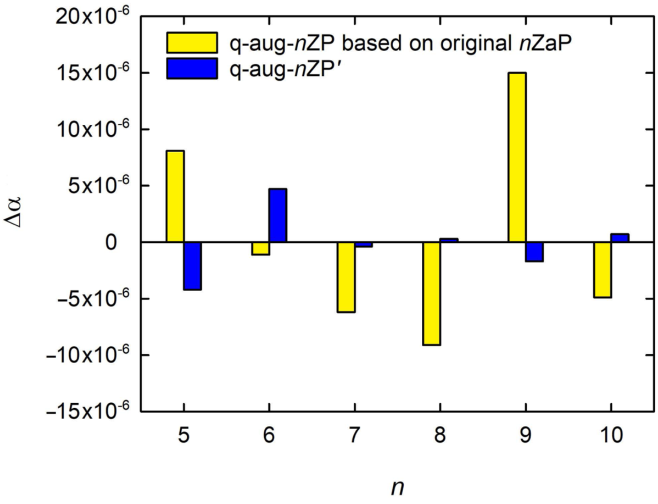

FIG. 3. Deviations of fitted values for the OS-MP2 polarizability contribution from the respective *ab initio* calculated values for the q-aug-nZP basis sets derived from the original nZaP basis sets [21] and for the q-aug-nZP′ basis sets derived from the reoptimized nZaP basis sets, nZaP′, as a function of n. The fits were performed with all n in the range from 5 to 10 using Eq. (8). Note that the final fit of the OS-MP2 polarizability contribution involves basis sets up to $n = 13$.

$$\Delta\alpha^{\mathrm{FC}}_{\mathrm{OS\text{-}MP2}} = 0.215\,897(8),$$

$$\Delta\alpha^{\mathrm{FC}}_{\mathrm{SS\text{-}MP2}} = 0.103\,976(1),$$

$$\Delta\alpha^{\mathrm{FC}}_{\mathrm{CCSD(T)-MP2}} = -0.033\,265(3),$$

where the given uncertainties were estimated from the stability of the extrapolated values upon varying the ranges of n values included in the extrapolation. Particularly for $\Delta\alpha^{\mathrm{FC}}_{\mathrm{OS\text{-}MP2}}$, it was crucial to use for up to $n = 10$ the reoptimized basis sets of this work in order to obtain a stable estimate for the CBS limit. Figure 3 illustrates how well Eq. (8) can fit the q-aug-nZP′ results for $n = 5$ to $n = 10$ in contrast to the respective q-aug-nZP results. The Supplemental Material [28] provides further details on the procedure to obtain the CBS estimate for $\Delta\alpha^{\mathrm{FC}}_{\mathrm{OS\text{-}MP2}}$ and its uncertainty.

The last term in Eq. (10), $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{CCSD(T)}}$, was computed using the aug-CnZP′, d-aug-CnZP′, and t-aug-CnZP′ basis sets with n ranging from 5 to 11 for the singly and doubly augmented basis sets and from 5 to 10 for the triply augmented ones. The resulting values are given in Table II. The extrapolation to the CBS limit was performed using Eq. (8) with the results for the basis sets d-aug-C7ZP′ to d-aug-C11ZP′, yielding

$$\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{CCSD(T)}} = -0.006\,976(3),$$

TABLE II. Calculated values for the polarizability difference between the AE-CCSD(T) and FC-CCSD(T) levels of theory, $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{CCSD(T)}}$, and the final extrapolated value.

n	aug-CnZP′	d-aug-CnZP′	t-aug-CnZP′
5	−0.006172	−0.006784	−0.006810
6	−0.006467	−0.006884	−0.006896
7	−0.006674	−0.006928	−0.006932
8	−0.006788	−0.006948	−0.006950
9	−0.006853	−0.006959	−0.006959
10	−0.006890	−0.006965	−0.006965
11	−0.006914	−0.006968	
∞		**−0.006976(3)**	

where, again, the uncertainty was estimated from the stability of the extrapolated value upon varying the range of values included in the fit of Eq. (8).

Adding the different contributions to $\alpha_{\mathrm{CCSD(T)}}$ and adding their uncertainties in quadrature, we obtain

$$\alpha_{\mathrm{CCSD(T)}} = 2.656\,376(9).$$

All contributions to $\alpha_{\mathrm{CCSD(T)}}$ were calculated using PSI3 [24].

Next, we discuss the correction for higher coupled-cluster levels up to FCI, $\Delta\alpha_{\mathrm{FCI-CCSD(T)}}$, which was split into a total of nine contributions,

$$\begin{aligned}\Delta\alpha_{\mathrm{FCI-CCSD(T)}} = {} & \Delta\alpha^{\mathrm{FC}}_{\mathrm{T-(T)}} + \Delta\alpha^{\mathrm{FC}}_{\mathrm{(Q)-T}} + \Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}} + \Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}} \\ & + \Delta\alpha^{\mathrm{FC}}_{\mathrm{H-P}} + \Delta\alpha^{\mathrm{FC}}_{\mathrm{FCI-H}} + \Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{(Q)-(T)}} \\ & + \Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{Q-(Q)}} + \Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{P-Q}}. \qquad (11)\end{aligned}$$

The terms $\Delta\alpha^{\mathrm{FC}}_{\mathrm{T-(T)}}$, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{(Q)-T}}$, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}}$, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}}$, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{H-P}}$, and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{FCI-H}}$ denote the polarizability differences within the FC approximation between the CCSDT [33] and CCSD(T), CCSDT(Q) [34,35] and CCSDT, CCSDTQ [35–37] and CCSDT(Q), CCSDTQP [38] and CCSDTQ, CCSDTQPH and CCSDTQP, and FCI and CCSDTQPH levels of theory, respectively. The contribution $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{(Q)-(T)}}$ corrects the terms $\Delta\alpha^{\mathrm{FC}}_{\mathrm{T-(T)}}$ and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{(Q)-T}}$ for the inclusion of the 1s electrons in the correlation treatment and was calculated by subtracting the polarizability difference between the FC-CCSDT(Q) and FC-CCSD(T) levels from the polarizability difference between the AE-CCSDT(Q) and AE-CCSD(T) levels. Similarly, $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{Q-(Q)}}$ and $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{P-Q}}$ are the AE corrections for $\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}}$ and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}}$. For $\Delta\alpha^{\mathrm{FC}}_{\mathrm{H-P}}$ and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{FCI-H}}$, the AE corrections could not be calculated with reasonably sized basis sets due to the extreme computational costs and, therefore, had to be neglected.

The contribution $\Delta\alpha^{\mathrm{FC}}_{\mathrm{T-(T)}}$ was calculated using the aug-nZP′ and d-aug-nZP′ basis sets up to $n = 9$ and the t-aug-nZP′ basis sets up to $n = 8$ (see Table III for the results). The general coupled-cluster code MRCC [39,40] was used for the calculations. The extrapolation to the CBS limit was performed using Eq. (7) with the d-aug-nZP′ basis sets using, as for all final extrapolations with this equation, the results for the two highest n values, which yields

$$\Delta\alpha^{\mathrm{FC}}_{\mathrm{T-(T)}} = -0.001\,039(2).$$

TABLE III. Calculated values for the polarizability difference between the FC-CCSDT and FC-CCSD(T) levels of theory, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{T-(T)}}$, and the final extrapolated value.

n	aug-nZP′	d-aug-nZP′	t-aug-nZP′
5	−0.000543	−0.000733	−0.000735
6	−0.000750	−0.000853	−0.000856
7	−0.000880	−0.000919	−0.000924
8	−0.000948	−0.000958	−0.000962
9	−0.000985	−0.000982	
∞		**−0.001039(2)**	

When the results for the second and third highest n values are used in the extrapolation, the resulting value for the CBS limit differs only by 1.3×10^{-6}, which justifies the given uncertainty estimate.

For the calculation of the term $\Delta\alpha^{\mathrm{FC}}_{\mathrm{(Q)-T}}$, we used CFOUR [29]. Since the public version of CFOUR can only handle basis functions up to i symmetry, the (d-,t-)aug-nZP′ basis sets can only be used for $n \leqslant 6$. To obtain larger basis sets that do not contain functions of k and higher symmetry, we removed the basis functions with the three highest angular momenta from the (d-,t-)aug-nZP′ basis sets up to $n = 9$. These basis sets are denoted as (d-,t-)aug-n_{-3}ZP′. Table IV lists the resulting values for $\Delta\alpha^{\mathrm{FC}}_{\mathrm{(Q)-T}}$. The final extrapolated value,

$$\Delta\alpha^{\mathrm{FC}}_{\mathrm{(Q)-T}} = 0.001\,162(5),$$

was obtained using the results for t-aug-8_{-3}ZP′ and t-aug-9_{-3}ZP′ with Eq. (7), in which n was replaced by $n - 3$ to account for the fact that this extrapolation scheme, on theoretical grounds, is not actually tied to the value of n but to the value of the highest angular momentum present in the basis set, l_{max}, which is normally identical to n. If the extrapolation is performed using the results for the basis sets t-aug-7_{-3}ZP′ and t-aug-8_{-3}ZP′, the resulting CBS estimate is 0.001 157, and the difference between the two CBS values is taken as the final value's uncertainty estimate.

The contributions $\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}}$, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}}$, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{H-P}}$, and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{FCI-H}}$ were calculated using (d-, t-)aug-cc-pVnZ basis sets, with the size of the largest basis set that could be handled with the available computational equipment steadily decreasing from $\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}}$ to $\Delta\alpha^{\mathrm{FC}}_{\mathrm{FCI-H}}$ (see Table V). The CCSDT(Q) and

TABLE IV. Calculated values for the polarizability difference between the FC-CCSDT(Q) and FC-CCSDT levels of theory, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{(Q)-T}}$, and the final extrapolated value. The employed (d-,t-)aug-n_{-3}ZP′ basis sets are identical to the (d-,t-)aug-nZP′ basis sets, except that the functions with the highest three angular momenta have been removed.

n	aug-n_{-3}ZP′	d-aug-n_{-3}ZP′	t-aug-n_{-3}ZP′
5	0.000558	0.000736	0.000746
6	0.000854	0.001032	0.001038
7	0.000992	0.001108	0.001110
8	0.001053	0.001132	0.001133
9	0.001091	0.001145	0.001145
∞			**0.001162(5)**

TABLE V. Calculated and final extrapolated values for the polarizability differences between the FC-CCSDTQ and FC-CCSDT(Q) levels of theory, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}}$; between the FC-CCSDTQP and FC-CCSDTQ levels of theory, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}}$; between the FC-CCSDTQPH and FC-CCSDTQP levels of theory, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{H-P}}$; and between the FC-FCI and FC-CCSDTQPH levels of theory, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{FCI-H}}$. Note the additional digit given for $\Delta\alpha^{\mathrm{FC}}_{\mathrm{FCI-H}}$ because of its very small magnitude.

n	aug-cc-pVnZ	d-aug-cc-pVnZ	t-aug-cc-pVnZ
		$\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}}$	
2	−0.000132	−0.000345	−0.000383
3	−0.000207	−0.000438	−0.000456
4	−0.000208	−0.000355	−0.000357
5	−0.000256	−0.000330	−0.000331
6	−0.000274	−0.000316	
∞		**−0.000297(7)**	
		$\Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}}$	
2	−0.000051	−0.000309	−0.000323
3	−0.000271	−0.000498	−0.000501
4	−0.000382	−0.000476	
∞		**−0.000460(16)**	
		$\Delta\alpha^{\mathrm{FC}}_{\mathrm{H-P}}$	
2	−0.000010	−0.000034	−0.000035
3	−0.000021	−0.000039	
∞		**−0.000042(2)**	
		$\Delta\alpha^{\mathrm{FC}}_{\mathrm{FCI-H}}$	
2	−0.0000003	−0.0000013	**−0.0000013(7)**
3	−0.0000006		

CCSDTQ calculations were performed with CFOUR, while all post-CCSDTQ calculations, including the FCI calculations, were carried out with MRCC. For $\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}}$, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}}$, and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{H-P}}$, the final extrapolated values were obtained using Eq. (7) with the results for the d-aug-cc-pVnZ basis sets,

$$\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}} = -0.000\,297(7),$$

$$\Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}} = -0.000\,460(16),$$

$$\Delta\alpha^{\mathrm{FC}}_{\mathrm{H-P}} = -0.000\,042(2),$$

where the uncertainty for $\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}}$ is the difference between the extrapolated value obtained with $n = 5$ and $n = 6$ and that obtained with $n = 4$ and $n = 5$, while for $\Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}}$ and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{H-P}}$ the uncertainties were estimated as the differences between the extrapolated values and those obtained with the largest basis sets. The FCI calculations could only be performed with the basis sets aug-cc-pVDZ, aug-cc-pVTZ, d-aug-cc-pVDZ, and t-aug-cc-pVDZ. Because the singly augmented basis sets are clearly not saturated with diffuse functions, we did not use them to extrapolate to the CBS limit. Instead, we used the value for the t-aug-cc-pVDZ basis set as our final result,

$$\Delta\alpha^{\mathrm{FC}}_{\mathrm{FCI-H}} = -1.3(7) \times 10^{-6},$$

where the uncertainty was estimated as one-half of the absolute value of this correction.

TABLE VI. Calculated and final extrapolated values for the AE corrections $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{(Q)-(T)}}$, $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{Q-(Q)}}$, and $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{P-Q}}$ of the contributions $\Delta\alpha^{\mathrm{FC}}_{\mathrm{(Q)-(T)}} = \Delta\alpha^{\mathrm{FC}}_{\mathrm{T-(T)}} + \Delta\alpha^{\mathrm{FC}}_{\mathrm{(Q)-T}}$, $\Delta\alpha^{\mathrm{FC}}_{\mathrm{Q-(Q)}}$, and $\Delta\alpha^{\mathrm{FC}}_{\mathrm{P-Q}}$, respectively.

n	aug-u-cc-pCVnZ	d-aug-u-cc-pCVnZ	t-aug-u-cc-pCVnZ
		$\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{(Q)-(T)}}$	
2	0.000007	0.000021	0.000020
3	−0.000013	−0.000033	−0.000033
4	−0.000012	−0.000027	−0.000027
5	−0.000012	−0.000022	−0.000022
6	−0.000014	−0.000020	−0.000020
∞			**−0.000018(3)**
		$\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{Q-(Q)}}$	
2	−0.000001	−0.000002	−0.000003
3	0.000002	0.000005	0.000005
4	0.000006	0.000008	0.000009
5	0.000008	0.000009	
∞		**0.000010(1)**	
		$\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{P-Q}}$	
2	0.000002	0.000005	0.000005
3	0.000008	0.000015	
∞		**0.000019(4)**	

The AE correction terms $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{(Q)-(T)}}$, $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{Q-(Q)}}$, and $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{P-Q}}$ were computed using (d-, t-)aug-u-cc-pCVnZ basis sets (see Table VI). The calculations up to the CCSDTQ level of theory were carried out with CFOUR, while for the CCSDTQP calculations the MRCC program was used. The CBS extrapolations were performed using Eq. (7) with the results for the triply augmented basis sets with $n = 5$ and $n = 6$ for $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{(Q)-(T)}}$ and with the results for the doubly augmented sets with $n = 4$ and $n = 5$ for $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{Q-(Q)}}$ and with $n = 2$ and $n = 3$ for $\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{P-Q}}$, yielding

$$\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{(Q)-(T)}} = -0.000\,018(3),$$

$$\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{Q-(Q)}} = 0.000\,010(1),$$

$$\Delta\alpha^{\mathrm{AE-FC}}_{\mathrm{P-Q}} = 0.000\,019(4),$$

where the uncertainty estimates were obtained as the differences between the extrapolated values and those for the largest basis sets.

The resulting final value for the FCI correction to $\alpha_{\mathrm{CCSD(T)}}$ is

$$\Delta\alpha_{\mathrm{FCI-CCSD(T)}} = -0.000\,667(19),$$

with which we obtain an FCI estimate of

$$\alpha_{\mathrm{FCI}} = 2.655\,710(21).$$

C. Finite nuclear mass and size corrections

In this work, the correction for finite nuclear mass effects, $\Delta\alpha_{\mathrm{FNM}}$, is the diagonal Born-Oppenheimer correction (DBOC) to the polarizability, which accounts for both mass scaling and mass polarization effects. The DBOC for the total energy of a neon atom can be written as [41,42]

$$\Delta E_{\mathrm{DBOC}} = -\frac{1}{2M_{\mathrm{nuc}}}\langle\Psi|\nabla^2_{\mathrm{nuc}}|\Psi\rangle, \tag{12}$$

TABLE VII. Calculated values for the DBOC at the AE-CCSD level of theory, $\Delta\alpha_{\rm DBOC}^{\rm CCSD}$; the DBOC difference between the AE-CCSDT and AE-CCSD levels, $\Delta\alpha_{\rm DBOC}^{\rm T-D}$; and the DBOC difference between the AE-CCSDTQ and AE-CCSDT levels, $\Delta\alpha_{\rm DBOC}^{\rm Q-T}$. Note the additional digit given for $\Delta\alpha_{\rm DBOC}^{\rm Q-T}$ because of its very small magnitude.

n	aug-u-cc-pCVnZ	d-aug-u-cc-pCVnZ	t-aug-u-cc-pCVnZ
		$\Delta\alpha_{\rm DBOC}^{\rm CCSD}$	
2	0.000020	0.000087	0.000090
3	0.000048	0.000082	0.000081
4	0.000068	0.000082	0.000082
5	0.000076	0.000082	0.000082
6	0.000078	0.000082	**0.000082(1)**
		$\Delta\alpha_{\rm DBOC}^{\rm T-D}$	
2	−0.000003	−0.000007	−0.000007
3	−0.000006	−0.000008	−0.000009
4	−0.000007	−0.000008	−0.000008
5	−0.000008	**−0.000008(1)**	
		$\Delta\alpha_{\rm DBOC}^{\rm Q-T}$	
2	−0.0000004	−0.0000016	−0.0000016
3	−0.0000012	**−0.0000022(6)**	

where $M_{\rm nuc}$ is the mass of the nucleus, Ψ is the normalized electronic wave function resulting with the Born-Oppenheimer approximation, and the operator $\nabla_{\rm nuc}^2$ acts on the nuclear coordinates. The DBOC to the polarizability is obtained as $\Delta\alpha_{\rm DBOC} = -(\partial^2\Delta E_{\rm DBOC}/\partial\mathscr{E}^2)_{\mathscr{E}=0}$ by applying, as before, the numerical differentiation approach described in Sec. III A.

We calculated $\Delta\alpha_{\rm FNM}$ as a sum of three DBOC terms,

$$\Delta\alpha_{\rm FNM} = \Delta\alpha_{\rm DBOC}^{\rm CCSD} + \Delta\alpha_{\rm DBOC}^{\rm T-D} + \Delta\alpha_{\rm DBOC}^{\rm Q-T}, \tag{13}$$

where $\Delta\alpha_{\rm DBOC}^{\rm CCSD}$ is the DBOC contribution at the AE-CCSD level of theory, $\Delta\alpha_{\rm DBOC}^{\rm T-D}$ denotes the DBOC difference between the AE-CCSDT and AE-CCSD levels, and $\Delta\alpha_{\rm DBOC}^{\rm Q-T}$ is the DBOC difference between the AE-CCSDTQ and AE-CCSDT levels. The three contributions were calculated using (d-, t-)aug-u-cc-pCVnZ basis sets. All calculations were performed for the dominant ^{20}Ne isotope, and the results were corrected by scaling them with the ratio of the mass of ^{20}Ne to the average atomic mass of natural neon. The scaled results are given in Table VII. Because the basis set convergence is somewhat erratic, we did not extrapolate these terms to the CBS limit. Instead, we used the values for the highest basis set levels (t-aug-u-cc-pCV6Z for $\Delta\alpha_{\rm DBOC}^{\rm CCSD}$, d-aug-u-cc-pCV5Z for $\Delta\alpha_{\rm DBOC}^{\rm T-D}$, and d-aug-u-cc-pCVTZ for $\Delta\alpha_{\rm DBOC}^{\rm Q-T}$), which are

$$\Delta\alpha_{\rm DBOC}^{\rm CCSD} = 0.000\,082(1),$$
$$\Delta\alpha_{\rm DBOC}^{\rm T-D} = -0.000\,008(1),$$
$$\Delta\alpha_{\rm DBOC}^{\rm Q-T} = -2.2(6)\times 10^{-6},$$

where the uncertainty of $\Delta\alpha_{\rm DBOC}^{\rm Q-T}$ was estimated as the difference between the values obtained with the d-aug-u-cc-pCVDZ and d-aug-u-cc-pCVTZ basis sets. From these results, we obtain

$$\Delta\alpha_{\rm FNM} = 0.000\,072(2).$$

To verify the correctness of our approach, we applied it also to helium. The $\Delta\alpha_{\rm FNM}$ value for ^{4}He that we obtained by calculating the DBOC at the FCI level with a triply augmented 7Z-quality basis set differs from the sum of the respective mass scaling contribution (which is a simple function of the ratio of the mass of an electron to that of the nucleus) and the mass polarization contribution determined by Pachucki and Sapirstein [7] by only about 0.1%.

While the DBOC calculations at the CCSD level of theory were performed with CFOUR [29,30], the CCSDT and CCSDTQ levels required the use of the interface between CFOUR and MRCC [39,40].

The finite nuclear size correction $\Delta\alpha_{\rm FNS}$ is extremely small. It was determined by Lesiuk *et al.* [10] to be only 1.4×10^{-6}. However, their value has the wrong sign, which is obvious from the equation they used to determine this contribution (which is correct). We adopt the corrected value for the present work,

$$\Delta\alpha_{\rm FNS} = -1.4(1)\times 10^{-6}.$$

D. Relativistic and QED corrections

All polarizability calculations reported in this paper so far correspond to the nonrelativistic limit, i.e., an infinite speed of light c. For a finite speed of light, the polarizability can be expanded in powers of $1/c$, and we can then write it in the form

$$\alpha = \alpha^{(0)} + \Delta\alpha_{\rm rel}^{(2)} + \Delta\alpha_{\rm QED}^{(3)} + \Delta\alpha_{\rm rel+QED}^{(4)} + \cdots, \tag{14}$$

where $\alpha^{(0)}$ is the nonrelativistic limit, $\Delta\alpha_{\rm rel}^{(2)}$ is the relativistic correction of order $1/c^2$, $\Delta\alpha_{\rm QED}^{(3)}$ is the QED contribution of order $1/c^3$, $\Delta\alpha_{\rm rel+QED}^{(4)}$ contains both relativistic and QED contributions of order $1/c^4$, and so forth. We calculated the total relativistic correction $\Delta\alpha_{\rm rel}$ as the sum of the $\Delta\alpha_{\rm rel}^{(2)}$ contribution and a correction $\Delta\alpha_{\rm rel}^{(4+)}$ for fourth- and higher-order contributions,

$$\Delta\alpha_{\rm rel} = \Delta\alpha_{\rm rel}^{(2)} + \Delta\alpha_{\rm rel}^{(4+)}, \tag{15}$$

while the QED correction $\Delta\alpha_{\rm QED}$ accounts for contributions up to order $1/c^4$,

$$\Delta\alpha_{\rm QED} = \Delta\alpha_{\rm QED}^{(3)} + \Delta\alpha_{\rm QED}^{(4)}. \tag{16}$$

For all relativistic and QED contributions considered in this work, the nucleus is approximated as a point of infinite mass. Thus, any finite nuclear mass and size effects are neglected.

The relativistic correction $\Delta\alpha_{\rm rel}^{(2)}$ follows from the Breit-Pauli Hamiltonian and is given for a closed-shell system, such as the neon atom, as

$$\Delta\alpha_{\rm rel}^{(2)} = \Delta\alpha_{\rm MV} + \Delta\alpha_{\rm D1} + \Delta\alpha_{\rm D2} + \Delta\alpha_{\rm SS} + \Delta\alpha_{\rm OO}, \tag{17}$$

where $\Delta\alpha_{\rm MV}$ is the mass-velocity term, $\Delta\alpha_{\rm D1}$ and $\Delta\alpha_{\rm D2}$ are the one- and two-electron Darwin contributions, $\Delta\alpha_{\rm SS}$ is the spin-spin term, and $\Delta\alpha_{\rm OO}$ is the orbit-orbit contribution. The spin-spin term can be eliminated for a closed-shell system using the relation $\Delta\alpha_{\rm SS} = -2\Delta\alpha_{\rm D2}$, so that we can write

$$\Delta\alpha_{\rm rel}^{(2)} = \Delta\alpha_{\rm MV} + \Delta\alpha_{\rm D1} - \Delta\alpha_{\rm D2} + \Delta\alpha_{\rm OO}. \tag{18}$$

The four polarizability contributions in Eq. (18) were obtained by applying again the numerical differentiation approach

TABLE VIII. Calculated and final extrapolated values for the sum of the mass-velocity and one-electron Darwin corrections, $\Delta\alpha_{\mathrm{MV}}^{\mathrm{CCSD(T)}} + \Delta\alpha_{\mathrm{D1}}^{\mathrm{CCSD(T)}}$, and the two-electron Darwin correction $\Delta\alpha_{\mathrm{D2}}^{\mathrm{CCSD(T)}}$ at the AE-CCSD(T) level of theory. The (d-, t-)aug-Cn_{-3}ZP′ basis sets are formed from the (d-, t-)aug-CnZP′ basis sets by removing the functions with the highest three angular momenta.

n	aug-Cn_{-3}ZP′	d-aug-Cn_{-3}ZP′	t-aug-Cn_{-3}ZP′
		$\Delta\alpha_{\mathrm{MV}}^{\mathrm{CCSD(T)}} + \Delta\alpha_{\mathrm{D1}}^{\mathrm{CCSD(T)}}$	
5	0.003952	0.004399	0.004424
6	0.004052	0.004362	0.004373
7	0.004126	0.004320	0.004324
8	0.004178	0.004304	0.004306
9	0.004211	0.004297	0.004298
∞			**0.004287(1)**
		$\Delta\alpha_{\mathrm{D2}}^{\mathrm{CCSD(T)}}$	
5	−0.000053	−0.000066	−0.000067
6	−0.000048	−0.000055	−0.000056
7	−0.000039	−0.000043	−0.000043
8	−0.000035	−0.000037	−0.000037
9	−0.000032	−0.000033	−0.000033
∞			−**0.000013(7)**

described in Sec. III A. Explicit expressions for the Hamiltonians from which ΔE_{MV}, ΔE_{D1}, ΔE_{D2}, and ΔE_{OO} are obtained can be found, for example, in Ref. [43].

The terms $\Delta\alpha_{\mathrm{MV}}$, $\Delta\alpha_{\mathrm{D1}}$, and $\Delta\alpha_{\mathrm{D2}}$ were calculated as

$$\Delta\alpha_{\mathrm{MV}} = \Delta\alpha_{\mathrm{MV}}^{\mathrm{CCSD(T)}} + \Delta\alpha_{\mathrm{MV}}^{\mathrm{T-(T)}} + \Delta\alpha_{\mathrm{MV}}^{\mathrm{Q-T}}, \tag{19}$$

$$\Delta\alpha_{\mathrm{D1}} = \Delta\alpha_{\mathrm{D1}}^{\mathrm{CCSD(T)}} + \Delta\alpha_{\mathrm{D1}}^{\mathrm{T-(T)}} + \Delta\alpha_{\mathrm{D1}}^{\mathrm{Q-T}}, \tag{20}$$

$$\Delta\alpha_{\mathrm{D2}} = \Delta\alpha_{\mathrm{D2}}^{\mathrm{CCSD(T)}} + \Delta\alpha_{\mathrm{D2}}^{\mathrm{T-(T)}} + \Delta\alpha_{\mathrm{D2}}^{\mathrm{Q-T}}, \tag{21}$$

where $\Delta\alpha_{\mathrm{MV}}^{\mathrm{CCSD(T)}}$ is the mass-velocity contribution at the AE-CCSD(T) level of theory, $\Delta\alpha_{\mathrm{MV}}^{\mathrm{T-(T)}}$ and $\Delta\alpha_{\mathrm{MV}}^{\mathrm{Q-T}}$ are the corrections for higher coupled-cluster levels up to AE-CCSDT and AE-CCSDTQ, respectively, and the individual terms for $\Delta\alpha_{\mathrm{D1}}$ and $\Delta\alpha_{\mathrm{D2}}$ have analogous definitions. All CCSD(T) and CCSDT calculations were performed using CFOUR [29,30], while the interface between CFOUR and MRCC was employed for the CCSDTQ calculations.

For the CCSD(T) calculations, we used (d-, t-)aug-Cn_{-3}ZP′ basis sets, which were obtained from the (d-, t-)aug-CnZP′ basis sets by removing the basis functions with the highest three angular momenta. The calculations were performed up to $n = 9$; the results are shown in Table VIII. Since there is always a large degree of cancellation between the mass-velocity and one-electron Darwin terms, it is advantageous to deal only with their sum and extrapolate this sum to the CBS limit. We performed the extrapolation using Eq. (7) (with n replaced by $n - 3$) with the triply augmented basis sets, yielding

$$\Delta\alpha_{\mathrm{MV}}^{\mathrm{CCSD(T)}} + \Delta\alpha_{\mathrm{D1}}^{\mathrm{CCSD(T)}} = 0.004\,287(1).$$

The extrapolation yields the same value within the given number of digits even if we use only the results for the basis sets with $n = 7$ and $n = 8$ or $n = 6$ and $n = 7$, so that the low uncertainty estimate is reasonable. For the CBS extrapolation of the two-electron Darwin term, we have to take into account that in this case the basis set incompleteness error is approximately proportional to $1/l_{\mathrm{max}}$ [44], so that we need to modify Eq. (7) by not only replacing n by $n - 3$ but also by changing the exponent in the denominator from 3 to 1. The extrapolated value is

$$\Delta\alpha_{\mathrm{D2}}^{\mathrm{CCSD(T)}} = -0.000\,013(7),$$

where the large uncertainty estimate (one-half of the absolute value of this term) was chosen to account for the slow basis set convergence, although it is reassuring that the extrapolation with $n = 7$ and $n = 8$ yields the same value within the given number of digits.

The corrections for the higher coupled-cluster levels were calculated using (d-, t-)aug-u-cc-pCVnZ basis sets. In the case of the terms $\Delta\alpha_{\mathrm{MV}}^{\mathrm{T-(T)}}$, $\Delta\alpha_{\mathrm{D1}}^{\mathrm{T-(T)}}$, and $\Delta\alpha_{\mathrm{D2}}^{\mathrm{T-(T)}}$, the calculations were performed up to $n = 6$ for all augmentation levels, whereas the terms $\Delta\alpha_{\mathrm{MV}}^{\mathrm{Q-T}}$, $\Delta\alpha_{\mathrm{D1}}^{\mathrm{Q-T}}$, and $\Delta\alpha_{\mathrm{D2}}^{\mathrm{Q-T}}$ could only be calculated up to $n = 4$ for the singly and doubly augmented sets and up to $n = 3$ for the triply augmented ones. The results are listed in Table IX. The CBS extrapolations for these terms were performed in the same way as for the corresponding terms at the AE-CCSD(T) level of theory (but taking into account that $l_{\mathrm{max}} = n$ instead of $l_{\mathrm{max}} = n - 3$), using for the AE-CCSDT corrections the values for the t-aug-u-cc-pCV5Z and t-aug-u-cc-pCV6Z basis sets and for the AE-CCSDTQ corrections the values for the d-aug-u-cc-pCVTZ and d-aug-u-cc-pCVQZ basis sets. The resulting final values are

$$\begin{aligned} \Delta\alpha_{\mathrm{MV}}^{\mathrm{T-(T)}} + \Delta\alpha_{\mathrm{D1}}^{\mathrm{T-(T)}} &= -0.000\,015(1), \\ \Delta\alpha_{\mathrm{D2}}^{\mathrm{T-(T)}} &= 4.0(3) \times 10^{-6}, \\ \Delta\alpha_{\mathrm{MV}}^{\mathrm{Q-T}} + \Delta\alpha_{\mathrm{D1}}^{\mathrm{Q-T}} &= 0.000\,012(3), \\ \Delta\alpha_{\mathrm{D2}}^{\mathrm{Q-T}} &= -2.6(6) \times 10^{-6}, \end{aligned}$$

where we obtain the same values for the AE-CCSDT corrections within the given number of digits when we extrapolate with the t-aug-u-cc-pCVQZ and t-aug-u-cc-pCV5Z basis sets. For $\Delta\alpha_{\mathrm{D2}}^{\mathrm{T-(T)}}$, the given uncertainty is the difference between the final value and that obtained by using the basis sets t-aug-u-cc-pCVTZ and t-aug-u-cc-pCVQZ for the extrapolation. For the AE-CCSDTQ corrections, the uncertainty estimates were obtained as the differences between the final values and those resulting from extrapolations with the d-aug-u-cc-pCVDZ and d-aug-u-cc-pCVTZ basis sets. We did not attempt to determine corrections for coupled-cluster levels beyond CCSDTQ because the CCSDTQ calculations were already extremely costly in terms of computing time.

We are not aware of any widely distributed quantum chemistry codes that could be used to compute the full orbit-orbit contribution $\Delta\alpha_{\mathrm{OO}}$. Lesiuk *et al.* [10], who refer to this contribution as the Breit correction, calculated it at the AE-CCSD(T) level of theory using an in-house code, and we adopt their value and uncertainty estimate for it,

$$\Delta\alpha_{\mathrm{OO}} = 0.001\,279(10).$$

The resulting total relativistic correction of order $1/c^2$ is

$$\Delta\alpha_{\mathrm{rel}}^{(2)} = 0.005\,575(12).$$

TABLE IX. Calculated and final extrapolated values for the difference between the sum of the mass-velocity and one-electron Darwin contributions obtained at the AE-CCSDT level of theory and the sum of these contributions obtained at the AE-CCSD(T) level, $\Delta\alpha_{\rm MV}^{\rm T-(T)} + \Delta\alpha_{\rm D1}^{\rm T-(T)}$, and for the difference between the two-electron Darwin contributions obtained at the AE-CCSDT and AE-CCSD(T) levels of theory, $\Delta\alpha_{\rm D2}^{\rm T-(T)}$. Furthermore, the analogously defined terms for the differences resulting from calculations at the AE-CCSDTQ and AE-CCSDT levels of theory, $\Delta\alpha_{\rm MV}^{\rm Q-T} + \Delta\alpha_{\rm D1}^{\rm Q-T}$ and $\Delta\alpha_{\rm D2}^{\rm Q-T}$, are listed. Note the additional digit given for $\Delta\alpha_{\rm D2}^{\rm T-(T)}$ and $\Delta\alpha_{\rm D2}^{\rm Q-T}$ because of their very small magnitude.

n	aug-u-cc-pCVnZ	d-aug-u-cc-pCVnZ	t-aug-u-cc-pCVnZ
		$\Delta\alpha_{\rm MV}^{\rm T-(T)} + \Delta\alpha_{\rm D1}^{\rm T-(T)}$	
2	0.000004	0.000003	0.000004
3	−0.000003	−0.000007	−0.000007
4	−0.000009	−0.000012	−0.000012
5	−0.000012	−0.000013	−0.000013
6	−0.000014	−0.000014	−0.000014
∞			**−0.000015(1)**
		$\Delta\alpha_{\rm D2}^{\rm T-(T)}$	
2	0.0000004	0.0000008	0.0000008
3	0.0000009	0.0000014	0.0000014
4	0.0000016	0.0000020	0.0000020
5	0.0000022	0.0000024	0.0000024
6	0.0000025	0.0000027	0.0000027
∞			**0.0000040(3)**
		$\Delta\alpha_{\rm MV}^{\rm Q-T} + \Delta\alpha_{\rm D1}^{\rm Q-T}$	
2	0.000000	0.000001	0.000001
3	0.000005	0.000007	0.000007
4	0.000009	0.000010	
∞		**0.000012(3)**	
		$\Delta\alpha_{\rm D2}^{\rm Q-T}$	
2	−0.0000003	−0.0000006	−0.0000006
3	−0.0000007	−0.0000010	−0.0000010
4	−0.0000012	−0.0000014	
∞		**−0.0000026(6)**	

TABLE X. Calculated values for the higher-order relativistic correction $\Delta\alpha_{\rm rel}^{(4+)}$ at the AE-CCSD(T) level of theory and the final extrapolated value. The correction was determined for a given basis set by taking the difference between the polarizability obtained with the 4cDC Hamiltonian and the sum of the respective nonrelativistic polarizability and the contributions $\Delta\alpha_{\rm MV}$, $\Delta\alpha_{\rm D1}$, and $\Delta\alpha_{\rm D2}$.

n	aug-u-cc-pCVnZ	d-aug-u-cc-pCVnZ	t-aug-u-cc-pCVnZ
2	0.000031	0.000065	0.000068
3	0.000045	0.000079	0.000079
4	0.000069	0.000082	0.000082
5	0.000074	0.000080	0.000080
6	0.000076	0.000080	0.000079
∞			**0.000078(26)**

The correction for higher-order relativistic effects, $\Delta\alpha_{\rm rel}^{(4+)}$, was determined by calculating the polarizability with the four-component Dirac-Coulomb (4cDC) Hamiltonian and subtracting the nonrelativistic polarizability and the contributions of order $1/c^2$ accounted for by the 4cDC Hamiltonian, which are $\Delta\alpha_{\rm MV}$, $\Delta\alpha_{\rm D1}$, and $\Delta\alpha_{\rm D2}$. Even though the 4cDC Hamiltonian accounts for relativistic contributions up to infinite order, the fact that it does not even account for all contributions of order $1/c^2$ shows that the treatment of higher-order relativistic corrections with this Hamiltonian is also inherently incomplete. Nevertheless, the sum of the terms $\Delta\alpha_{\rm MV}$, $\Delta\alpha_{\rm D1}$, and $\Delta\alpha_{\rm D2}$ accounts for about 76% of the value of $\Delta\alpha_{\rm rel}^{(2)}$, so that we assume that the majority of higher-order effects is accounted for by the 4cDC Hamiltonian. We calculated $\Delta\alpha_{\rm rel}^{(4+)}$ at the AE-CCSD(T) level of theory with the (d-, t-)aug-u-cc-pCVnZ basis sets up to $n = 6$ for all augmentation levels (see Table X for the results). The extrapolation to the CBS limit was performed using Eq. (7) with the results for the basis sets t-aug-u-cc-pCV5Z and t-aug-u-cc-pCV6Z, yielding

$$\Delta\alpha_{\rm rel}^{(4+)} = 0.000\,078(26),$$

where the relatively large uncertainty (one-third of the total value of $\Delta\alpha_{\rm rel}^{(4+)}$) was chosen to account for the described shortcomings of our approach. The 4cDC calculations were carried out using the DIRAC code [45,46] with explicit calculation of the two-electron (SS|SS) integrals over the small component.

For the total relativistic correction $\Delta\alpha_{\rm rel}$, we then obtain

$$\Delta\alpha_{\rm rel} = 0.005\,653(29).$$

To determine the QED correction of order $1/c^3$, we used a simplified expression similar to that applied by Lesiuk *et al.* [10], but with an additional contribution from the two-electron Darwin term (see, e.g., Ref. [9]),

$$\Delta\alpha_{\rm QED}^{(3)} = \frac{8}{3\pi c}\left(\frac{19}{30} + 2\ln c - \ln k_0\right)\Delta\alpha_{\rm D1} - \frac{1}{\pi c}\left(\frac{164}{15} - \frac{14}{3}\ln c\right)\Delta\alpha_{\rm D2}, \tag{22}$$

where $\ln k_0$ is the Bethe logarithm for the neon atom, for which we adopted the value of Lesiuk *et al.* [10], $\ln k_0 = 7.595$, which was calculated at the SCF level of theory and has a stated uncertainty of 1–2%. Compared with the full expression for $\Delta\alpha_{\rm QED}^{(3)}$ given in Ref. [9], Eq. (22) neglects the electric-field dependence of the Bethe logarithm and the Araki-Sucher contribution. Furthermore, as already mentioned, we neglect any corrections to the QED contributions for finite nuclear mass effects. However, at least in the case of helium it is known that none of these three simplifications affect the total QED correction of order $1/c^3$ by more than 1% [8,9]. The value for the one-electron Darwin correction $\Delta\alpha_{\rm D1}$ in Eq. (22) was obtained in the same way as the sum $\Delta\alpha_{\rm MV} + \Delta\alpha_{\rm D1}$ by extrapolating the contribution at the AE-CCSD(T) level of theory and the AE-CCSDT and AE-CCSDTQ corrections to the CBS limit. The resulting final value for $\Delta\alpha_{\rm D1}$ is $-0.017\,349(13)$. For $\Delta\alpha_{\rm D2}$, the final value is $-0.000\,012(7)$. This yields

$$\Delta\alpha_{\rm QED}^{(3)} = -0.000\,310(62),$$

where the uncertainty estimate of 20% of the total value of $\Delta\alpha_{\rm QED}^{(3)}$ is adopted from Lesiuk *et al.* [10]. The contribution

TABLE XI. Summary of the main contributions to the polarizability of neon obtained in this work and by Lesiuk *et al.* [10] as well as the respective final values and the experimental value of Gaiser and Fellmuth [4] (for which the given uncertainty is the standard uncertainty).

Contribution	This work	Lesiuk *et al.* (Ref. [10])	Gaiser and Fellmuth (Ref. [4])
FCI polarizability α_{FCI}	2.655710(21)	2.65545(35)	
Finite nuclear mass correction $\Delta\alpha_{\mathrm{FNM}}$	0.000072(2)	0.00022(1)	
Finite nuclear size correction $\Delta\alpha_{\mathrm{FNS}}$	−0.0000014(1)	0.0000014(1)	
Relativistic correction $\Delta\alpha_{\mathrm{rel}}$	0.005653(29)	0.00550(5)	
QED correction $\Delta\alpha_{\mathrm{QED}}$	−0.000366(68)	−0.00037(7)	
Total polarizability α	2.661067(77)	2.66080(36)	2.661057(7)

due to the two-electron Darwin term accounts only for about 0.1% of the value of $\Delta\alpha^{(3)}_{\mathrm{QED}}$.

The QED contribution of order $1/c^4$ is approximated by the one-loop correction, which is given as [10]

$$\Delta\alpha^{(4)}_{\mathrm{QED}} = \frac{2Z}{c^2}\left(\frac{427}{96} - 2\ln 2\right)\Delta\alpha_{\mathrm{D1}}, \tag{23}$$

where $Z = 10$ is the nuclear charge of neon. The resulting value of the correction is

$$\Delta\alpha^{(4)}_{\mathrm{QED}} = -0.000\,057(28),$$

where we adopted the uncertainty estimate of Lesiuk *et al.* [10] of 50% of the absolute value of $\Delta\alpha^{(4)}_{\mathrm{QED}}$, which also accounts for the neglected higher-order QED contributions.

The resulting total QED correction is

$$\Delta\alpha_{\mathrm{QED}} = -0.000\,366(68),$$

which together with α_{FCI}, $\Delta\alpha_{\mathrm{FNM}}$, $\Delta\alpha_{\mathrm{FNS}}$, and $\Delta\alpha_{\mathrm{rel}}$ gives a final polarizability of

$$\alpha = 2.661\,067(77).$$

IV. DISCUSSION AND CONCLUSIONS

The main contributions to the static electric-dipole polarizability of this work and the resulting total value are compared in Table XI with the respective values obtained by Lesiuk *et al.* [10] and with the experimental polarizability obtained by Gaiser and Fellmuth [4]. Further theoretical and experimental values for neon's polarizability can be found in the literature (see, e.g., the review on the polarizabilities of the elements by Schwerdtfeger and Nagle [47] and Table XI of Lesiuk *et al.*'s paper [10]). However, because of the high accuracy of the theoretical value of Lesiuk *et al.* [10] and the experimental value measured by Gaiser and Fellmuth using DCGT, we compare the present results only with the values from these two groups.

Our value for the polarizability in the nonrelativistic FCI limit assuming a point nucleus of infinite mass, α_{FCI}, is consistent with the corresponding value obtained by Lesiuk *et al.* [10] within their uncertainty estimate, but the present value is about an order of magnitude more accurate. This contribution could be further improved in a straightforward manner by using even larger basis sets for the calculations, but this appears to be economically unfeasible at the moment.

In the case of the finite nuclear mass correction $\Delta\alpha_{\mathrm{FNM}}$, our result is in severe disagreement with that of Lesiuk *et al.* [10]. The reason for this disagreement is that Lesiuk *et al.* considered only the effect of mass scaling and neglected mass polarization effects (which are included in our DBOC treatment of $\Delta\alpha_{\mathrm{FNM}}$). Lesiuk *et al.* [10] justified their approach by using helium as an example, for which Pachucki and Sapirstein [7] showed that mass polarization accounts for only about 8% of the total finite nuclear mass effect. However, mass polarization in helium is only a post-SCF effect, whereas for neon it is already present at the SCF level due to the occupied *p* orbitals and is thus much larger.

As already mentioned, Lesiuk *et al.* [10] determined the finite nuclear size effect $\Delta\alpha_{\mathrm{FNS}}$ with the wrong sign. However, due to the very small magnitude of $\Delta\alpha_{\mathrm{FNS}}$, this mistake is of no consequence.

The values for the relativistic correction of this work and that of Lesiuk *et al.* [10] do not agree within mutual uncertainties, but the agreement is still reasonable. The present value is more accurate because Lesiuk *et al.* [10] treated all relativistic effects only at the AE-CCSD(T) level of theory, whereas we also included corrections for higher coupled-cluster levels up to AE-CCSDTQ for the mass-velocity and one- and two-electron Darwin contributions. Moreover, our relativistic correction includes an estimate for contributions of order $1/c^4$ and higher, which is missing in the work of Lesiuk *et al.* [10].

The QED corrections of this work and of Lesiuk *et al.* [10] are, within rounding errors, identical. This is due to the fact that we adopted their value for the Bethe logarithm and because the values for the only other quantity that significantly influences the QED corrections, the one-electron Darwin term, differ by only about 0.3%.

The total polarizability value of this work agrees with that of Lesiuk *et al.* [10] within the latter's uncertainty, but the present uncertainty, which is only about 0.003% of the total value, is almost five times smaller than that of Lesiuk *et al.* The comparison of our result with the experimental value of Gaiser and Fellmuth [4] shows a remarkable level of agreement; we agree with their value almost within their standard uncertainty, which is an order of magnitude smaller than the uncertainty estimate of our value. In contrast, the value of Lesiuk *et al.* [10] differs from Gaiser and Fellmuth's value by about a factor of 27 more than the present value. While the very close agreement between our polarizability and that of Gaiser and Fellmuth [4] is certainly to some extent due to a fortuitous cancellation of errors, it could also indicate that our uncertainty estimate might be too conservative. Since our overall uncertainty budget is dominated by the uncertainty of the QED corrections, which we adopted from Lesiuk *et al.* [10], this could mean that the uncontrolled approximations that had to be made when calculating the QED corrections are not severe. Furthermore, the uncertainty of the higher-order relativistic corrections makes a considerable contribution to the overall uncertainty budget because our approximate treatment of these contributions at the 4cDC level also constitutes an uncontrolled approximation. Ultimately, the question to which degree the close agreement between our polarizability value and that of Gaiser and Fellmuth [4] is fortuitous can only be resolved by a more complete treatment of QED and higher-order relativistic effects, and we hope that this paper will stimulate further research in this direction.

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Correction: The value given for α_{exp} in the penultimate sentence of the abstract was set incorrectly during the proof production process and has been fixed.