Nuclear magnetic shielding in heliumlike ions

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Ab initio QED calculations of the nuclear magnetic shielding constant in heliumlike ions are presented. We combine the nonrelativistic QED approach based on an expansion in powers of the fine-structure constant α and the so-called all-order QED approach, which includes all orders in the parameter $Z\alpha$ but uses a perturbation expansion in the parameter $1/Z$ (where Z is the nuclear charge number). The combination of the two complementary methods makes our treatment applicable to both low-Z and high-Z ions. Our calculations confirm the presence of a rare antiscreening effect for the relativistic shielding correction and demonstrate the importance of the inclusion of the negative-energy part of the Dirac spectrum.

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I. INTRODUCTION

Magnetic moments of nuclei are often determined from nuclear magnetic resonance (NMR) measurements. Despite the high precision of these experiments, the accuracy of the extracted nuclear moments is severely limited by the restricted knowledge of the magnetic shielding caused by the chemical surrounding. Such effects are difficult to calculate reliably, which has often led to significant deficiencies in the literature data on nuclear magnetic moments [1,2]. As an example, the so-called bismuth hyperfine puzzle [3,4] was recently resolved [5] and traced back to an inaccuracy of the nuclear magnetic moment caused by shortcomings in calculations of the shielding correction.

Much more accurate determinations of nuclear magnetic moments can be achieved by Penning-trap measurements of the combined Zeeman and hyperfine structure of few-electron atoms or ions. The shielding constants of such systems can be calculated *ab initio* within the framework of QED, with a detailed analysis of uncertainties due to omitted higher-order effects. Precise determinations of magnetic moments of a number of light nuclei by this method were reported in recent years [6–9]. In particular, the magnetic moment of the proton was accurately measured by the Penning-trap technique in Ref. [10]. This technique can in principle be extended to measurements of other nuclei and closed-shell ions.

Highly sophisticated calculations of the nuclear shielding have been recently accomplished for the helium atom [11–13], motivated by perspectives of using the hyperpolarized helium NMR probes as a new standard for absolute magnetometry [14–16]. The calculation of Ref. [11] revealed a rare effect of antiscreening for the relativistic shielding correction, corresponding to the situation when an effect for two correlated $(1s)^2$ electrons is larger than for two noninteracting $1s$ electrons. This defies the physically intuitive picture in which each of the correlated electrons should experience a slightly smaller nuclear charge because it is effectively screened by the second electron, the effect commonly known as screening. So far the presence of the antiscreening effect has not been confirmed by an independent calculation.

The goal of the present study is to perform *ab initio* QED calculations of the nuclear magnetic shielding of heliumlike ions for a wide range of nuclear charges Z. This is achieved by merging together two complementary methods, namely, the nonrelativistic QED (NRQED) approach based on an expansion in powers of the fine-structure constant α and the so-called all-order QED approach, which includes all orders in the parameter $Z\alpha$ but uses a perturbation expansion in the parameter $1/Z$. The NRQED method alone is applicable only to low-Z ions, since the uncalculated higher-order effects scale with high powers of Z. By contrast, the all-order method is effective in the high-Z region, since the $1/Z$ expansion converges fast there. In this work we unify these two methods so that the resulting approach becomes applicable for the whole range of Z. For the first time such a unified approach was applied by Drake to calculate energies and transition rates of heliumlike ions in Refs. [17,18].

The outline of our calculations is as follows. First, we employ the NRQED approach to calculate the leading shielding contribution of order α^2 as well as the relativistic, nuclear, and QED corrections of orders α^4, $\alpha^2 m/M$, and $\alpha^5 \ln\alpha$. Then we address the higher-order corrections within the all-order method. We calculate the one-electron shielding contribution, the one-photon exchange, QED, and the nuclear magnetization distribution effects. By analyzing the $Z\alpha$ expansion of the individual corrections, we identify the lowest-order contributions already included in the NRQED treatment and remove them, thus avoiding double counting and obtaining the final results for the shielding constant.

II. NONRELATIVISTIC QED APPROACH

Within the NRQED approach, the shielding constant σ of low-Z atoms is represented as a double expansion in α and the electron-to-nucleus mass ratio m/M,

$$\sigma = \alpha^2\sigma^{(2)} + \alpha^4\sigma^{(4)} + \alpha^2\frac{m}{M}\sigma^{(2,1)} + \alpha^5\sigma^{(5)} + \cdots . \quad (1)$$

As is customary in NRQED calculations, we will use atomic units in the following formulas in this section. To the leading order in α and zeroth order in m/M, the shielding constant σ takes the form [19]

$$\sigma^{(2)} = \frac{1}{3}\sum_a\left\langle\frac{1}{r_a}\right\rangle, \quad (2)$$

where r_a is the distance between the nucleus and ath electron and the summation runs over all electrons. The relativistic shielding correction of order α^4 was derived in Ref. [11], with the result

$$\begin{aligned}\sigma^{(4)} = {} & \sum_a\left\langle\frac{1}{12r_a^3}\left(\frac{\vec{r}\cdot\vec{r}_1\,\vec{r}\cdot\vec{r}_2}{r^3} - 3\frac{\vec{r}_1\cdot\vec{r}_2}{r}\right) - \frac{1}{6}\left(\frac{1}{r_a}p_a^2 + \frac{(\vec{r}_a\times\vec{p}_a)^2}{r_a^3} + 4\pi\,\delta(\vec{r}_a)\right)\right\rangle \\ & + \frac{2}{3}\left\langle\left(\frac{1}{r_1}+\frac{1}{r_2}\right)\frac{1}{(E-H)'}\left[\sum_a\left(\frac{\pi Z}{2}\delta(\vec{r}_a) - \frac{p_a^4}{8}\right) + \pi\,\delta(\vec{r}) - \frac{1}{2}p_1^i\left(\frac{\delta^{ij}}{r} + \frac{r^i r^j}{r^2}\right)p_2^j\right]\right\rangle \\ & - \frac{2}{9}\left\langle\pi[\delta(\vec{r}_1)-\delta(\vec{r}_2)]\frac{1}{E-H}\left(3p_1^2 - 3p_2^2 - \frac{Z}{r_1} + \frac{Z}{r_2} - \frac{\vec{r}\cdot(\vec{r}_1+\vec{r}_2)}{r^3}\right)\right\rangle \\ & - \frac{1}{6}\left\langle\left(\frac{\vec{r}_1\times\vec{p}_1}{r_1^3} + \frac{\vec{r}_2\times\vec{p}_2}{r_2^3}\right)\frac{1}{E-H}\left(\vec{r}_1\times\vec{p}_1p_1^2 + \vec{r}_2\times\vec{p}_2p_2^2 + \frac{1}{r}\vec{r}_1\times\vec{p}_2 + \frac{1}{r}\vec{r}_2\times\vec{p}_1 - \vec{r}_1\times\vec{r}_2\frac{\vec{r}}{r^3}\cdot(\vec{p}_1+\vec{p}_2)\right)\right\rangle \\ & - \frac{1}{8}\left\langle\left(\frac{r_1^ir_1^j}{r_1^5} - \frac{r_2^ir_2^j}{r_2^5}\right)^{(2)}\frac{1}{E-H}\left(Z\frac{r_1^ir_1^j}{r_1^3} - Z\frac{r_2^ir_2^j}{r_2^3} + \frac{r^i}{r^3}(r_1^j + r_2^j)\right)^{(2)}\right\rangle, \end{aligned} \quad (3)$$

where $\vec{r}\equiv\vec{r}_1-\vec{r}_2$, $(p^iq^i)^{(2)} = p^iq^j/2 + p^jq^i/2 - \delta^{ij}\vec{p}\cdot\vec{q}/3$, and $1/(E-H)'$ is the reduced Green's function (with the reference state removed from the sum over the spectrum).

The leading-order nuclear recoil correction was derived in Ref. [20] and later corrected in Ref. [13]. The result is

$$\begin{aligned}\sigma^{(2,1)} &\equiv \frac{1-g_N}{g_N}\sigma_a^{(2,1)} + \sigma_b^{(2,1)} \\ &= \frac{1-g_N}{g_N}\frac{\langle p_N^2\rangle}{3Z} + \frac{1}{3}\left\langle\sum_a\frac{1}{r_a}\frac{1}{(E-H)'}p_N^2\right\rangle \\ &\quad + \frac{1}{3}\left\langle\sum_a\vec{r}_a\times\vec{p}_N\frac{1}{E-H}\sum_b\frac{\vec{r}_b}{r_b^3}\times\vec{p}_b\right\rangle, \end{aligned} \quad (4)$$

where $\vec{p}_N = -\sum_a\vec{p}_a$; $g_N = (M/m_p)(\mu/\mu_N)/(ZI)$ is the nuclear g factor; M, I, and μ are the nuclear mass, spin, and magnetic moment, respectively; m_p is the proton mass; and μ_N is the nuclear magneton.

The logarithmic part of the leading QED correction of order α^5 was derived in Ref. [11]. We write it as

$$\begin{aligned}\sigma_{\log}^{(5)} = \ln(Z\alpha)\Bigg(&-\frac{16Z}{9}\left\langle\sum_a\frac{1}{r_a}\frac{1}{(E-H)'}\sum_b\delta(\vec{r}_b)\right\rangle \\ &+ \frac{28}{9}\left\langle\sum_a\frac{1}{r_a}\frac{1}{(E-H)'}\delta(\vec{r}\,)\right\rangle - \frac{40}{9}\left\langle\sum_a\delta(\vec{r}_a)\right\rangle\Bigg). \end{aligned} \quad (5)$$

This formula differs from the one from Ref. [11] by $\ln Z$ in the second-order correction containing $\delta(\vec{r}\,)$. In obtaining it, we take into account that the two-electron Lamb shift contains, in addition to $\ln\alpha$, an implicit $\ln Z$ term usually hidden in the Araki-Sucher term $\langle r^{-3}\rangle$ [21]. The nonlogarithmic QED contribution of order α^5 was derived and calculated for helium in Ref. [12]. Its numerical calculation is rather complicated as it involves perturbations of the so-called Bethe logarithm. For this reason, we address this and higher-order QED corrections within the $1/Z$ expansion in the next section.

Numerical calculations of the NRQED corrections summarized above were carried out with the basis set of exponential functions $e^{-\alpha_ir_1-\beta_ir_2-\gamma_ir}$ [22]. The method of calculations was developed in our previous investigations; see Ref. [23] for a review. The most computationally intense part was the calculation of $\sigma^{(4)}$. While the evaluation of first-order matrix elements in Eq. (3) was relatively straightforward, the computation of second-order matrix elements turned out to be rather demanding. To achieve high numerical accuracy, we used carefully optimized basis sets for the intermediate electron states. The optimization was carried out for symmetric second-order corrections, using nonuniform distributions of nonlinear basis-set parameters (see Ref. [23] for details), with a typical size of the basis set $N = 1200$. The obtained basis sets were then used for computation of nonsymmetric second-order corrections in Eq. (3).

Results of our numerical calculations of $\sigma^{(2)}$, $\sigma^{(2,1)}$, $\sigma^{(4)}$, and $\sigma_{\log}^{(5)}$ for $Z \leqslant 12$ are presented in Table I. The corresponding values for $Z > 12$ can be readily obtained by using the $1/Z$ expansion, which is of the form

$$\frac{\sigma^{(2)}}{Z} = \sum_{k=0}^{\infty}\frac{c_k}{Z^k}, \quad (6)$$

and similarly for other corrections. The leading coefficients c_0 are known analytically from the hydrogen theory, whereas

TABLE I. NRQED shielding corrections for different nuclear charges Z and their $1/Z$-expansion coefficients c_k.

Z	$\sigma^{(2)}/Z$	$\sigma^{(4)}/Z^3$	$\sigma^{(5)}_{\log}/[Z^3\ln(Z\alpha)]$	$\sigma^{(2,1)}_a/Z$	$\sigma^{(2,1)}_b/Z$
2	0.562 772 266 9	2.321 754 4	−0.710 693 3	0.510 465 64	−0.597 289 84
	0.562 772 266 8[a]	2.321 42[a]			
3	0.597 316 533	2.070 397 4	−0.837 418 1	0.560 658 46	−0.619 929 38
4	0.614 625 068	1.979 393 5	−0.906 430 9	0.586 503 61	−0.631 434 60
5	0.625 021 856	1.933 018 3	−0.949 438 3	0.602 232 65	−0.638 394 84
6	0.631 957 258	1.905 101 6	−0.978 729 3	0.612 807 06	−0.643 058 72
7	0.636 912 900	1.886 523 9	−0.999 940 0	0.620 402 23	−0.646 401 72
8	0.640 630 484	1.873 300 7	−1.016 000 2	0.626 121 09	−0.648 915 36
9	0.643 522 387	1.863 423 1	−1.028 579 1	0.630 582 19	−0.650 874 23
10	0.645 836 163	1.855 771 1	−1.038 696 3	0.634 159 23	−0.652 443 72
11	0.647 729 404	1.849 672 8	−1.047 009 0	0.637 091 24	−0.653 729 45
12	0.649 307 201	1.844 700 8	−1.053 960 0	0.639 538 22	−0.654 801 99
c_0	$\frac{2}{3}$	$\frac{97}{54}$	$-\frac{32}{9\pi}$	$\frac{2}{3}$	$-\frac{2}{3}$
c_1	$-\frac{5}{24}$	0.514 442 6	0.947 740	−0.327 804	0.143 12
c_2	0.000 000 0	0.770 0	−0.159 8	0.026 44	−0.008 9
c_3	0.002 899 7	0.288	−0.104	0.008 6	0.000 3
c_4	−0.000 592	0.40	0.003		
c_5	−0.001 04				

[a]Reference [11].

the higher-order coefficients c_k were obtained by fitting our numerical results.

For the relativistic correction $\sigma^{(4)}$ we find a small deviation from the helium result of Ref. [11]. The difference comes from the second-order contribution with the ${}^3D$ intermediate states, labeled as Q_{12} in Ref. [11]. The deviation is small and does not influence the final theoretical prediction for the helium shielding constant within its estimated error.

Our results summarized in Table I confirm the previous findings [11] of the presence of the unusual antiscreening effect for $\sigma^{(4)}$. Indeed, the absolute values of $\sigma^{(4)}/Z^3$ for all nuclear charges are larger than the corresponding limiting value for noninteracting electrons, $c_0 = \frac{97}{54} \approx 1.796$. This is in contrast to all other corrections examined in Table I, which exhibit the normal screening effect. It is important to note that the first two $1/Z$-expansion coefficients c_0 and c_1 of $\sigma^{(4)}$ are independently cross-checked by our calculation of the one-photon-exchange correction in Sec. III, thus excluding the possibility of a technical mistake in the derivation of $\sigma^{(4)}$. The probable explanation of the antiscreening effect is the singlet-triplet mixing. Specifically, the relativistic effects mix the ground $1\,{}^1S_0$ state with intermediate $n\,{}^3S$ states. This mixing is quite large and changes the behavior of the relativistic correction in the case of heliumlike atoms compared to the hydrogenlike case.

III. ALL-ORDER APPROACH

In order to access the higher-order effects of order α^{5+}, we will adopt the so-called all-order QED approach. This method includes all orders in the parameter $Z\alpha$ but expands in the electron-electron interaction, with the expansion parameter $1/Z$. In order to separate out the higher-order contributions beyond what is already included into the NRQED treatment in Sec. II, we examine the $Z\alpha$ expansion of the all-order results and remove the double counting by subtracting the leading-order contributions. The zeroth order in $1/Z$ is delivered by the independent-particle approximation, which neglects the interaction between the electrons. Further terms of the $1/Z$ expansion are described by Feynman diagrams containing an exchange of one, two, etc., virtual photons between the electrons. In this section we will use relativistic units $\hbar = c = 1$.

A. Electron-structure effects

We start by examining the so-called electron-structure effects, which are induced by Feynman diagrams without radiative loops.

1. One-electron case

In the independent-particle approximation, the relativistic shielding constant for the $(1s)^2$ state of the heliumlike ion is (see Ref. [24] for details)

$$\sigma_{\rm rel,\,1el} = \alpha \sum_{\mu_a} \sum_{n\neq a} \frac{1}{\varepsilon_a - \varepsilon_n} \langle a|V_g|n\rangle\langle n|V_h|a\rangle, \tag{7}$$

where μ_a is the momentum projection of the $1s$ electron, the sum over n runs over the complete spectrum of the Dirac equation, and V_g and V_h are effective interactions of a Dirac electron with the external magnetic field V_g and with the magnetic dipole nuclear field V_h,

$$V_g = (\vec{r}\times\vec{\alpha})_z, \quad V_h = \frac{(\vec{r}\times\vec{\alpha})_z}{r^3}. \tag{8}$$

For the point nuclear charge, Eq. (7) can be calculated analytically [25–29], with the result

$$\sigma_{\rm rel,1el} = 2\left(-\frac{4\alpha Z\alpha}{9}\right)\left(\frac{1}{3} - \frac{1}{6(1+\gamma)} + \frac{2}{\gamma} - \frac{3}{2\gamma-1}\right) = 2\alpha Z\alpha\left(\frac{1}{3} + \frac{97}{108}(Z\alpha)^2 + \frac{289}{216}(Z\alpha)^4 + \frac{3269}{1728}(Z\alpha)^6 + \cdots\right), \tag{9}$$

where $\gamma = \sqrt{1-(Z\alpha)^2}$. For an extended nuclear charge distribution, Eq. (7) can be readily calculated numerically with help of the dual-kinetic-balance finite-basis-set method [30].

The higher-order one-electron relativistic correction is obtained from the expression (9) by subtracting the first two terms of the $Z\alpha$ expansion,

$$\sigma^{\rm HO}_{\rm rel,1el} = \sigma_{\rm rel,1el} - 2\alpha(Z\alpha)\left[\tfrac{1}{3} + \tfrac{97}{108}(Z\alpha)^2\right]. \tag{10}$$

It should be noted that when performing the summation over the Dirac energy spectrum in Eq. (7), the inclusion of the negative-energy part of the spectrum is mandatory, as its contribution is very large, especially for low-Z ions. This is explained by the fact that the nonrelativistic limit of the nuclear shielding constant in atoms is induced solely by the negative-energy part of the Dirac spectrum. So for $Z=2$ the negative-energy states induce 99.9% of the total result. With the increase of Z, the relative contribution of the negative-energy states gradually diminishes but is still prominent, e.g., for $Z=60$ it is 37%. This is in sharp contrast to calculations of transition energies, where the negative-energy contribution is suppressed by a factor of $(Z\alpha)^3$ compared to the leading nonrelativistic result [31].

2. One-photon exchange

The one-photon exchange correction to the nuclear magnetic shielding can be obtained as a perturbation of the one-photon exchange correction to the energy by two external interactions V_g and V_h. For the ground state of a heliumlike ion, we obtain

$$\sigma_{\rm rel,1ph} = \alpha\sum_P(-1)^P[\langle PaPb|I|\delta_{hg}ab\rangle + \langle PaPb|I|\delta_{gh}ab\rangle + \langle PaPb|I|\delta_h a\delta_g b\rangle + \langle\delta_h PaPb|I|\delta_g ab\rangle + \langle\delta_h PaPb|I|a\delta_g b\rangle - \langle PaPb|I|ab\rangle\langle\delta_h a|\delta_g a\rangle - \langle a|V_g|a\rangle\langle PaPb|I|\tilde{\delta}_h ab\rangle - \langle a|V_h|a\rangle\langle PaPb|I|\tilde{\delta}_g ab\rangle], \tag{11}$$

where a and b denote the two electrons in the $(1s)^2$ shell, P is the permutation operator ($PaPb = ab$ or ba), $(-1)^P$ is the sign of the permutation, the perturbations of the wave functions are defined by

$$|\delta_i a\rangle = \sum_n^{\varepsilon_n\neq\varepsilon_a}\frac{|n\rangle\langle n|V_i|a\rangle}{\varepsilon_a-\varepsilon_n}, \tag{12}$$

$$|\tilde{\delta}_i a\rangle = \sum_n^{\varepsilon_n\neq\varepsilon_a}\frac{|n\rangle\langle n|V_i|a\rangle}{(\varepsilon_a-\varepsilon_n)^2}, \tag{13}$$

$$|\delta_{hg} a\rangle = \sum_{n_1}^{\varepsilon_{n_1}\neq\varepsilon_a}\sum_{n_2}^{\varepsilon_{n_2}\neq\varepsilon_a}\frac{|n_1\rangle\langle n_1|V_h|n_2\rangle\langle n_2|V_g|a\rangle}{(\varepsilon_a-\varepsilon_{n_1})(\varepsilon_a-\varepsilon_{n_2})}, \tag{14}$$

and $|\delta_{gh}a\rangle$ is obtained from Eq. (14) by interchanging g and h. Furthermore, $I \equiv I(r_{12})$ denotes the electron-electron interaction operator in the Coulomb gauge for the zero transferred energy,

$$I(r_{12}) = \frac{\alpha}{r_{12}} - \frac{\alpha}{2r_{12}}[\vec{\alpha}_1\cdot\vec{\alpha}_2 + (\vec{\alpha}_1\cdot\hat{\vec{r}}_{12})(\vec{\alpha}_2\cdot\hat{\vec{r}}_{12})], \tag{15}$$

where $\vec{\alpha}$ are the Dirac matrices, $\vec{r}_{12} = \vec{r}_1 - \vec{r}_2$, and $\hat{\vec{r}} = \vec{r}/r$. We note that in obtaining Eq. (11) we took into account that the two electrons in the $(1s)^2$ shell have the same energy, so the frequency dependence of the electron-electron interaction operator does not play any role in this case.

We calculated Eq. (11) numerically with help of the B-spline basis-set method [32] for the point-charge nuclear model and with the dual-kinetic-balance method [30] for the extended-charge nuclear model. The typical basis size used in the computation was $N = 100$–150. In order to achieve high numerical accuracy in the low-Z region (required for a high-precision fitting of the d_2 coefficient), we had to use quadruple precision (approximately 32 decimal digits) in our computation.

The results for the point nuclear model are listed in Table II. In order to remove the double counting with the NRQED results, we analyze the $Z\alpha$ expansion of the one-

TABLE II. One-photon exchange shielding correction $\sigma_{\rm rel,1ph}$ for different nuclear charges and coefficients of its $Z\alpha$ expansion, for the point nuclear model.

Z	$\sigma_{\rm rel,1ph}/\alpha^2$
2	−0.208 223 677
3	−0.208 086 391
4	−0.207 893 786
6	−0.207 340 881
8	−0.206 560 303
10	−0.205 545 466
14	−0.202 776 655
18	−0.198 940 386
22	−0.193 905 594
28	−0.183 713 434
34	−0.169 616 681
40	−0.150 469 629
46	−0.124 653 329
52	−0.089 862 182
d_0	$-\frac{5}{24}$
d_2	0.514 442 6
d_4	1.693 21
d_6	2.469

photon exchange correction, which is of the form

$$\sigma_{\mathrm{rel,1ph}} = \alpha^2 \sum_{k=0}^{\infty} d_{2k} (Z\alpha)^{2k}. \tag{16}$$

The coefficients d_{2k} obtained by fitting our numerical results are listed in Table II. The first coefficient $d_0 = -\frac{5}{24}$ corresponds to the $1/Z^1$ coefficient of expansion of $\sigma^{(2)}/Z$ (see Table I). The second coefficient d_2 corresponds to the $1/Z^1$ coefficient of $\sigma^{(4)}/Z^3$ (see Table I). Other coefficients represent contributions of order α^6 and higher which have not been accounted for in Sec. II. The higher-order one-photon exchange contribution is obtained as

$$\sigma^{\mathrm{HO}}_{\mathrm{rel,1ph}} = \sigma_{\mathrm{rel,1ph}} - \alpha^2 [d_0 + d_2 (Z\alpha)^2], \tag{17}$$

where d_0 and d_2 are listed in Table II.

Analyzing contributions induced by the positive- and negative-energy parts of the Dirac spectrum in the one-photon exchange shielding correction, we observe that, similarly to the one-electron case, the contribution of the negative-energy states is rather large. For $Z = 2$, they contribute 99.9% of the total result, whereas for $Z = 60$, the negative- and positive-energy contributions are of the same magnitude and of opposite sign. This demonstrates the importance of the proper treatment of the negative-energy part of the Dirac spectrum in calculations of the nuclear shielding. We note that a similar dominance of the negative-energy contribution was recently found for the $M1$ polarizability in strontium [33].

3. Exchange of two or more photons

The uncertainty due to the exchange of two or more photons is estimated based on the pattern of the available $1/Z$-expansion coefficients of the $\sigma^{(4)}$ correction. Specifically,

$$\sigma^{\mathrm{HO}}_{\mathrm{rel,2ph+}} \approx \pm \sigma^{\mathrm{HO}}_{\mathrm{rel,1ph}} 2 \frac{c_2}{c_1 Z} \approx \pm \sigma^{\mathrm{HO}}_{\mathrm{rel,1ph}} \frac{3}{Z}, \tag{18}$$

where 2 is the conservative factor.

B. QED effects

In the independent-particle approximation, the QED contribution for the $(1s)^2$ state is twice the $1s$ QED correction calculated for hydrogenlike ions to all orders in $Z\alpha$ in Refs. [24,34]. The analytical result for the leading $Z\alpha$-expansion term was obtained in Refs. [24,34] and later corrected in Ref. [12]. Separating out the leading-order result, we represent the one-electron QED contribution for the $(1s)^2$ state of heliumlike ions as

$$\sigma_{\mathrm{QED,1el}} = 2\alpha^2 (Z\alpha)^3 \left(-\frac{16}{9\pi} \ln(Z\alpha) - 1.896\,642\,389 + G_{\mathrm{QED}}(Z\alpha) \right), \tag{19}$$

where $G_{\mathrm{QED}}(Z\alpha) \approx 2.182(Z\alpha) + O((Z\alpha)^2)$ is the remainder function containing one-electron contributions of higher orders in $Z\alpha$. The logarithmic term in the formula (19) corresponds to the $1/Z^0$ term of the expansion of $\sigma^{(5)}_{\log}$, whereas the other terms have not been included in the NRQED treatment of Sec. II. The remainder function $G_{\mathrm{QED}}(Z\alpha)$ was calculated to all orders in $Z\alpha$ in Refs. [24,34]. In the present work we use the data obtained in those works and ascribe to it the relative uncertainty of $\pm 0.3(Z\alpha)^2$, to account for the uncalculated diagrams with magnetic-loop vacuum polarization. So the higher-order QED contribution beyond those included in Sec. II is

$$\sigma^{\mathrm{HO}}_{\mathrm{QED,1el}} = 2\alpha^2 (Z\alpha)^3 [-1.896\,642\,389 + G_{\mathrm{QED}}(Z\alpha)]. \tag{20}$$

In order to estimate the effects of the screening of the one-electron QED correction by the second electron, we introduce the screening factor ζ_{scr} basing on known results for the logarithmic QED contribution $\sigma^{(5)}_{\log}$. Specifically, we define

$$\zeta_{\mathrm{scr}} = -\frac{\sigma^{(5)}_{\log}/[Z^3 \ln(Z\alpha)] - c_0}{c_0}, \tag{21}$$

where $c_0 = -\frac{32}{9\pi}$ is the leading $1/Z$-expansion coefficient (see Table I). Using this screening factor, we estimate the QED screening contribution as

$$\sigma^{\mathrm{HO}}_{\mathrm{QED,1ph+}} \approx -\zeta_{\mathrm{scr}} \sigma^{\mathrm{HO}}_{\mathrm{QED,1el}} \pm 30\%. \tag{22}$$

This estimate of uncertainty is supported by the complete NRQED calculation of $\sigma^{(5)}$ for helium [12].

C. Nuclear magnetization distribution

Within the independent-particle approximation, the nuclear magnetization distribution correction for the $(1s)^2$ state is twice the $1s$ hydrogenlike contribution, derived to the leading order in $Z\alpha$ in Ref. [13]. Formulas presented in Ref. [13] include both the finite nuclear size (FNS) and the nuclear magnetization distribution [Bohr-Weisskopf (BW)] effects.

TABLE III. Individual contributions to the shielding constant in He-like ions, in units of 10^{-6}.

Contribution	$^{7}Li^{+}$	$^{9}Be^{2+}$	$^{17}O^{6+}$	$^{43}Ca^{18+}$	$^{73}Ge^{30+}$	$^{129}Xe^{52+}$
$\sigma^{(2)}$	95.423 74	130.918 47	272.9155	698.924	1124.94	1906.0
$\sigma^{(2,1)}$	−0.013 36	−0.020 13	−0.0229	−0.029	−0.04	−0.0
$\sigma^{(4)}$	0.158 52	0.359 23	2.7198	41.378	168.48	806.5
$\sigma^{(5)}_{\log}$	0.001 79	0.004 24	0.0306	0.346	1.09	3.4
$\sigma^{\mathrm{HO}}_{\mathrm{rel,1el}}$	0.000 04	0.000 23	0.0102	1.150	12.92	204.2
$\sigma^{\mathrm{HO}}_{\mathrm{rel,1ph+}}$	0.000 02 (2)	0.000 06 (5)	0.0010 (4)	0.041 (6)	0.29 (3)	2.7 (2)
$\sigma^{\mathrm{HO}}_{\mathrm{QED}}$	−0.001 53 (16)	−0.003 90 (29)	−0.0341 (12)	−0.544 (8)	−2.23 (4)	−11.4 (5)
$\sigma^{\mathrm{HO}}_{\mathrm{BW}}$	−0.000 02 (1)	−0.000 12 (6)	−0.0029 (14)	−0.148 (74)	−1.13 (57)	−10.8 (54)
σ	95.569 20 (16)	131.258 09 (30)	275.6172 (19)	741.119 (75)	1304.30 (57)	2900.5 (54)

TABLE IV. Nuclear magnetic shielding constant σ for the ground state of heliumlike ions with the nuclear charge number Z and mass number A.

Z	A	$\sigma \times 10^3$	Z	A	$\sigma \times 10^3$	Z	A	$\sigma \times 10^3$
3	7	0.095 569 20 (16)	30	67	1.200 56 (43)	56	137	3.106 4 (64)
4	9	0.131 258 09 (30)	31	69	1.251 86 (49)	57	139	3.214 8 (69)
5	11	0.167 087 88 (50)	32	73	1.304 30 (57)	58	139	3.327 0 (74)
6	13	0.203 069 60 (79)	33	75	1.358 04 (65)	59	141	3.443 3 (79)
7	14	0.239 245 6 (12)	34	77	1.413 05 (74)	60	143	3.563 5 (85)
8	17	0.275 617 2 (19)	35	79	1.469 43 (83)	62	149	3.816 9 (99)
9	19	0.312 259 7 (29)	36	83	1.527 20 (94)	63	151	3.951 (11)
10	21	0.349 167 0 (44)	37	85	1.586 5 (11)	64	155	4.089 (12)
11	23	0.386 411 0 (63)	38	87	1.647 3 (12)	65	159	4.235 (12)
12	25	0.423 972 7 (88)	39	89	1.709 8 (13)	66	161	4.382 (13)
13	27	0.461 945 (12)	40	91	1.774 0 (15)	67	165	4.539 (14)
14	29	0.500 306 (16)	41	93	1.839 9 (16)	68	167	4.699 (15)
15	31	0.539 126 (22)	42	95	1.907 6 (18)	69	169	4.869 (16)
17	35	0.618 236 (38)	43	99	1.977 3 (20)	70	171	5.043 (17)
18	39	0.658 585 (48)	44	101	2.049 0 (22)	71	175	5.222 (18)
19	39	0.699 566 (60)	45	103	2.122 9 (25)	72	177	5.414 (19)
20	43	0.741 119 (75)	46	105	2.199 0 (27)	73	181	5.611 (20)
21	45	0.783 380 (92)	47	107	2.277 4 (30)	74	183	5.817 (22)
22	47	0.826 32 (11)	48	111	2.358 2 (32)	75	185	6.033 (23)
23	51	0.870 04 (13)	49	113	2.441 6 (35)	76	187	6.255 (24)
24	53	0.914 52 (16)	50	119	2.527 6 (39)	77	191	6.489 (26)
25	55	0.959 85 (19)	51	121	2.616 3 (42)	78	195	6.731 (27)
26	57	1.006 07 (23)	52	125	2.707 9 (46)	79	197	6.985 (29)
27	59	1.053 18 (27)	53	127	2.802 6 (50)	80	199	7.07 (12)
28	61	1.101 27 (31)	54	129	2.900 5 (54)	83	209	8.102 (42)
29	63	1.150 39 (37)	55	133	3.001 7 (59)	91	231	10.92 (13)

Removing the FNS part, we get

$$\sigma_{\mathrm{BW,1el}} = 2\left(-\frac{2\alpha(Z\alpha)^3}{3}\right)\left[m^2 r_M^2 + 4Z\alpha m(r_Z - \langle r\rangle)\right], \quad (23)$$

where r_M and r_Z are the magnetic and the Zemach radius, respectively, and $\langle r\rangle$ is the mean nuclear charge radius. Within the Gaussian model for the nuclear charge distribution $\rho_C(r) = \rho_0 \exp(-3r^2/2r_C^2)$ (and similar to the magnetization distribution), we obtain

$$r_Z = \sqrt{\frac{8}{3\pi}}\sqrt{r_C^2 + r_M^2}, \quad \langle r\rangle = \sqrt{\frac{8}{3\pi}}r_C, \quad (24)$$

where $r_C = \langle r^2\rangle^{1/2}$ is the root-mean-square nuclear charge radius.

For light nuclei with $Z = 3$ and 4, we use Eq. (23) with the experimental values of the Zemach radii $r_Z(^7\mathrm{Li}) = 3.33$ fm [35] and $r_Z(^9\mathrm{Be}) = 4.04$ fm [36]. For heavier nuclei, the Zemach radius is not readily available from experiment. For some nuclei, the $1s$ shielding BW correction was calculated in Ref. [24] within the effective single-particle model of the nuclear magnetization distribution. However, this model is not universal and is applicable for some selected nuclei only. In the present work we use Eq. (23) with the magnetic radius expressed in terms of the charge radius by $r_M = \sqrt{3}r_C$ for nuclear charges $Z < 80$. We find that with this choice of the magnetic radius, Eq. (23) qualitatively reproduces results of the single-particle model calculations of Ref. [24]. We estimate the uncertainty of this approximation of the one-electron BW correction to be 50%, which can be compared to the 30% uncertainty estimate of the single-particle model results in Ref. [24]. For $Z > 80$, Eq. (23) is no longer adequate. We thus apply the single-particle nuclear model as described in Ref. [24] to compute the BW correction for several high-Z ions, specifically, with $Z = 80$, 83, and 91. The effects of the electron-electron interaction on the one-electron BW corrections are estimated analogously to Eq. (22).

IV. RESULTS AND DISCUSSION

In this work we performed numerical calculations of the nuclear shielding correction for the ground state of heliumlike ions for a wide range of nuclear charges. The nuclear parameters were taken from Ref. [2] (magnetic moments), Ref. [37] (charge radii), and Ref. [38] (masses). Individual shielding contributions for selected ions are presented in Table III. We observe that for the lightest ions, the dominant theoretical uncertainty comes from the QED screening effect. This uncertainty can be improved further by a calculation of the nonlogarithmic α^5 QED correction, as accomplished for helium in Ref. [12]. For heavier ions, the largest theoretical uncertainty comes from the extended distribution of the nuclear magnetic moment (the BW effect). This uncertainty can in principle be improved by dedicated calculations of the BW correction for specific nuclei with a microscopic nuclear model [39]. An even better way is to use experimental values of the effective Zemach radius $\tilde{r}_Z$ [35] obtained from the hyperfine-splitting measurements. One can then use $\tilde{r}_Z$ instead of r_Z in Eq. (23) and compute r_M from Eq. (24). An additional benefit is that this automatically accounts for some higher-order nuclear effects included in $\tilde{r}_Z$.

Table IV lists our theoretical predictions of the nuclear shielding constant for heliumlike ions. We do not present results for neutral helium since more complete calculations are available in this case [12,13]. The absolute accuracy of theoretical predictions for the shielding constant σ varies from 2×10^{-10} for $Z = 3$ to 1×10^{-4} for $Z = 91$. This accuracy demonstrates the precision possible for determination of nuclear magnetic moments from heliumlike ions.

Summarizing, we performed calculations of the nuclear magnetic shielding for heliumlike ions in the ground state. By combining two complementary approaches, we obtained results for a wide region of nuclear charges. Our calculations confirmed the presence of a rare antiscreening effect for the relativistic shielding correction. They also demonstrated the importance of inclusion of the negative-energy part of the Dirac spectrum in calculations of the nuclear shielding, especially for low-Z ions. In the future, the developed approach can be extended to calculations of nuclear shielding in Li-like ions, which are of immediate experimental interest [8,9,40].

[1] P. Raghavan, At. Data Nucl. Data Tables **42**, 189 (1989).
[2] N. J. Stone, At. Data Nucl. Data Tables **90**, 75 (2005).
[3] J. Ullmann, Z. Andelkovic, C. Brandau, A. Dax, W. Geithner, C. Geppert, C. Gorges, M. Hammen, V. Hannen, S. Kaufmann *et al.*, Nat. Commun. **8**, 15484 (2017).
[4] J.-P. Karr, Nat. Phys. **13**, 533 (2017).
[5] L. V. Skripnikov, S. Schmidt, J. Ullmann, C. Geppert, F. Kraus, B. Kresse, W. Nörtershäuser, A. F. Privalov, B. Scheibe, V. M. Shabaev, M. Vogel, and A. V. Volotka, Phys. Rev. Lett. **120**, 093001 (2018).
[6] M. Puchalski, J. Komasa, A. Spyszkiewicz, and K. Pachucki, Phys. Rev. A **105**, 042802 (2022).
[7] A. Schneider, B. Sikora, S. Dickopf, M. Müller, N. S. Oreshkina, A. Rischka, I. A. Valuev, S. Ulmer *et al.*, Nature (London) **606**, 878 (2022).
[8] K. Pachucki, V. Patkóš, and V. A. Yerokhin, Phys. Lett. B **846**, 138189 (2023).
[9] K. Pachucki and M. Puchalski, Opt. Commun. **283**, 641 (2010).
[10] A. Mooser, S. Ulmer, K. Blaum, K. Franke, H. Kracke, C. Leiteritz, W. Quint, C. C. Rodegheri, C. Smorra, and J. Walz, Nature (London) **509**, 596 (2014).
[11] A. Rudziński, M. Puchalski, and K. Pachucki, J. Chem. Phys. **130**, 244102 (2009).
[12] D. Wehrli, A. Spyszkiewicz-Kaczmarek, M. Puchalski, and K. Pachucki, Phys. Rev. Lett. **127**, 263001 (2021).
[13] K. Pachucki, Phys. Rev. A **108**, 062806 (2023).
[14] A. Nikiel, P. Blümler, W. Heil, M. Hehn, S. Karpuk, A. Maul, E. Otten, L. M. Schreiber, and M. Terekhov, Eur. Phys. J. D **68**, 330 (2014).
[15] X. Fan, S. E. Fayer, and G. Gabrielse, Rev. Sci. Instrum. **90**, 083107 (2019).
[16] M. Farooq, T. Chupp, J. Grange, A. Tewsley-Booth, D. Flay, D. Kawall, N. Sachdeva, and P. Winter, Phys. Rev. Lett. **124**, 223001 (2020).
[17] G. W. F. Drake, Phys. Rev. A **19**, 1387 (1979).
[18] G. W. F. Drake, Can. J. Phys. **66**, 586 (1988).
[19] N. F. Ramsey, Phys. Rev. **78**, 699 (1950).
[20] K. Pachucki, Phys. Rev. A **78**, 012504 (2008).
[21] G. W. F. Drake and A. J. Makowsky, J. Phys. B **18**, L103 (1985).
[22] V. I. Korobov, Phys. Rev. A **61**, 064503 (2000).
[23] V. A. Yerokhin, V. Patkóš, and K. Pachucki, Symmetry **13**, 1246 (2021).
[24] V. A. Yerokhin, K. Pachucki, Z. Harman, and C. H. Keitel, Phys. Rev. A **85**, 022512 (2012).
[25] E. A. Moore, Mol. Phys. **97**, 375 (1999).
[26] N. C. Pyper, Mol. Phys. **97**, 381 (1999).
[27] N. C. Pyper and Z. C. Zhang, Mol. Phys. **97**, 391 (1999).
[28] D. L. Moskovkin, N. S. Oreshkina, V. M. Shabaev, T. Beier, G. Plunien, W. Quint, and G. Soff, Phys. Rev. A **70**, 032105 (2004).
[29] V. G. Ivanov, S. G. Karshenboim, and R. N. Lee, Phys. Rev. A **79**, 012512 (2009).
[30] V. M. Shabaev, I. I. Tupitsyn, V. A. Yerokhin, G. Plunien, and G. Soff, Phys. Rev. Lett. **93**, 130405 (2004).
[31] J. Sucher, Phys. Rev. A **22**, 348 (1980).
[32] W. R. Johnson, S. A. Blundell, and J. Sapirstein, Phys. Rev. A **37**, 307 (1988).
[33] F.-F. Wu, T.-Y. Shi, W.-T. Ni, and L.-Y. Tang, Phys. Rev. A **108**, L051101 (2023).
[34] V. A. Yerokhin, K. Pachucki, Z. Harman, and C. H. Keitel, Phys. Rev. Lett. **107**, 043004 (2011).
[35] K. Pachucki, V. Patkóš, and V. A. Yerokhin, Phys. Rev. A **108**, 052802 (2023).
[36] M. Puchalski and K. Pachucki, Phys. Rev. A **89**, 032510 (2014).
[37] I. Angeli and K. Marinova, At. Data Nucl. Data Tables **99**, 69 (2013).
[38] M. Wang, W. Huang, F. G. Kondev, G. Audi, and S. Naimi, Chin. Phys. C **45**, 030003 (2021).
[39] I. Valuev, Ph.D. thesis, University of Heidelberg, 2022.
[40] S. Dickopf (private communication).