QED theory of the nuclear magnetic shielding in hydrogen-like ions

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The shielding of the nuclear magnetic moment by the bound electron in hydrogen-like ions is calculated *ab initio* with inclusion of relativistic, nuclear, and quantum electrodynamics (QED) effects. The QED correction is evaluated to all orders in the nuclear binding strength parameter and, independently, to the first order in the expansion in this parameter. The results obtained lay the basis for the high-precision determination of nuclear magnetic dipole moments from measurements of the g-factor of hydrogen-like ions.

PACS numbers: 31.30.jn, 31.15.ac, 32.10.Dk, 21.10.Ky

Magnetic dipole moments of nuclei are most often determined by the nuclear magnetic resonance (NMR) technique. Other methods such as atomic beam magnetic resonance, collinear laser spectroscopy, and optical pumping (OP) have also been used. The measured quantities are usually the ratio of the frequencies (or the g-factors) for the nucleus of interest and the reference nucleus. Such ratios can be experimentally determined with a part-per-billion (ppb) accuracy [1]. However, magnetic moments of *bare* nuclei extracted from these experiments are much less accurate. This is because the experimental data should be corrected for several physical effects, which are difficult to calculate. The main effect is the diamagnetic shielding of the external magnetic field by the electrons in the atom. The NMR results should be also corrected for the paramagnetic chemical shift caused by the chemical environment [2] and the OP data are sensitive to the hyperfine mixing of the energy levels [3]. Significant (and generally unknown) uncertainties of calculations of these effects often lead to ambiguities in the published values of nuclear magnetic moments [4].

Since the accuracy of calculations of the chemical shifts cannot be reliably assessed, the means of comparison of nuclear moments shielded by different environments in NMR measurements are rather limited. Independent determinations of nuclear magnetic moments would define uncertainties of theoretical calculations of the chemical shifts and help to assess the accuracy of NMR standards.

Reliable determination of the nuclear magnetic moments is also prompted by a new generation of QED calculations of the hyperfine splitting in highly charged ions. It was demonstrated [5] that the magnetic sector of bound-state QED can be tested in these systems to all orders in the binding field, if the nuclear magnetic moments are accurately known. Alternatively, comparing theoretical predictions with experimental results, one can determine nuclear properties and set benchmark tests for nuclear-structure theory. A recent example is the spectroscopic determination of the nuclear charge radii of the neutron-halo nuclei ^{8}He, ^{11}Li, and ^{11}Be [6], which yielded unique information about the properties of these extraordinary systems.

A way to a high-precision determination of nuclear magnetic moments is to study the simplest atomic systems, the hydrogen-like ions. Measurements of the bound-electron g-factor in these systems progressed dramatically during the recent years and reached the ppb level [7]. They led not only to a stringent test of sophisticated QED calculations [8, 9] but also to an improved determination of the electron mass [10]. Extensions of these experiments to ions with a nonzero nuclear spin will provide a determination of the nuclear magnetic moments from a simple system that can be described theoretically up to a very high accuracy.

It is well known [11] that the nuclear-spin-dependent part of the atomic g-factor g_F is suppressed by about 3 orders of magnitude as compared to the leading effect due to the bound-electron g-factor (see Eq. (1) below). This imposes limitations on possible determinations of the nuclear magnetic moment from g_F. We show here, however, that the leading effect cancels exactly in the sum of the g-factors for two hyperfine-structure levels (see Eq. (2) below). This sum is proportional to the nuclear g-factor and, therefore, is much better suited for extracting the nuclear magnetic moment. Its calculation can be conveniently parameterized in terms of the nuclear shielding constant σ, as given by Eq. (2).

In this work we perform an *ab initio* calculation of the nuclear magnetic shielding for the ground state of hydrogen-like ions. The relativistic, QED, and nuclear effects are accounted for. The main challenge is the calculation of the QED correction. To the best of our knowledge, the only attempt to address it was the estimate reported in Ref. [12]. In this Letter, we calculate the QED correction rigorously to all orders in the binding nuclear strength parameter $Z\alpha$ (where Z is the nuclear charge and α is the fine-structure constant) and, independently, we derive the leading term of its $Z\alpha$ expansion.

We now turn to the theory of the g-factor of a hydrogen-like ion with a nonzero spin. Within relativistic quantum mechanics, it is given by [11],

$$g_F^{(0)} = g_j\,\frac{\langle \boldsymbol{j}\cdot\boldsymbol{F}\rangle}{F(F+1)} - \frac{m}{m_p}\,g_I\,\frac{\langle \boldsymbol{I}\cdot\boldsymbol{F}\rangle}{F(F+1)}\,, \tag{1}$$

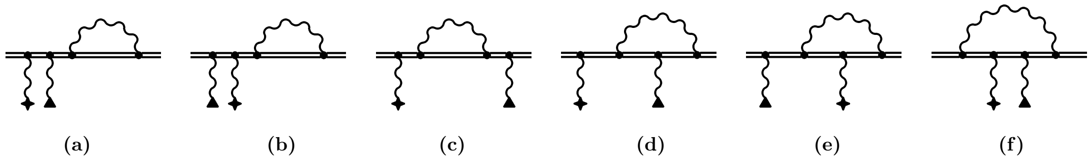

FIG. 1: Self-energy correction to the nuclear magnetic shielding. Double line represents the electron in the binding nuclear field. Wave line terminated by a triangle represents the dipole hyperfine interaction with the nucleus and wave line terminated by a cross represents the interaction with the external magnetic field.

where F is the total angular momentum, I is the nuclear spin, j is the electron angular momentum, g_j is the Dirac bound-electron g-factor, $g_I = \mu/(\mu_N I)$ is the nuclear g-factor, μ is the nuclear magnetic moment, $\mu_N = |e|/(2m_p)$ is the nuclear magneton, m and m_p are the electron and proton masses, respectively, $\langle \boldsymbol{j}\cdot\boldsymbol{F}\rangle = [F(F+1) - I(I+1) + j(j+1)]/2$, and $\langle \boldsymbol{I}\cdot\boldsymbol{F}\rangle = [F(F+1) + I(I+1) - j(j+1)]/2$. The higher-order corrections enter into Eq. (1) in two ways: (i) the Dirac electron g-factor g_j is modified by QED and recoil effects that do not depend on nuclear spin, (ii) the free-nucleus g-factor g_I is shielded by the bound electron. Additional corrections, e.g., those due to the electric quadrupole interaction [13], are small and can be absorbed into the definition of the nuclear shielding.

For the ground state of an ion with a nuclear spin $I > 1/2$, we introduce the combination of g-factors $\overline{g}$,

$$\overline{g} \equiv g_{F=I+1/2} + g_{F=I-1/2} = -2\frac{m}{m_p}\frac{\mu}{\mu_N I}(1-\sigma)\,, \quad (2)$$

whith σ being the shielding constant. If both g-factors $g_{F=I+1/2}$ and $g_{F=I-1/2}$ are measured and σ is known from theory, the above formula determines the nuclear magnetic moment μ. For the ions with a nuclear spin $I = 1/2$, Eq. (2) is not applicable and the nuclear magnetic moment has to be determined from Eq. (1).

The nuclear shielding constant σ defined by Eq. (2) can be represented as a sum

$$\sigma = \sigma^{(0)} + \delta\sigma_{\rm QED} + \delta\sigma_{\rm rec} + \delta\sigma_{\rm BW} + \delta\sigma_Q\,, \quad (3)$$

where $\sigma^{(0)}$ is the leading-order relativistic result (including the finite nuclear size effect), $\delta\sigma_{\rm QED}$ is the QED correction, $\delta\sigma_{\rm rec}$ is the recoil correction, $\delta\sigma_{\rm BW}$ is the nuclear magnetization distribution (Bohr-Weisskopf) correction, and $\delta\sigma_Q$ is the electric quadrupole correction.

The exact relativistic result for the leading-order magnetic shielding $\sigma^{(0)}$ was obtained analytically (for a point nucleus) [14] and numerically [13]. The recoil correction is known [12] to the leading order in $Z\alpha$,

$$\delta\sigma_{\rm rec} = -\frac{\alpha Z\alpha}{3}\frac{m}{M}\left(1 + \frac{g_N - 1}{g_N}\right)\,, \quad (4)$$

where M is the nuclear mass and $g_N = M\mu/(\mu_N\, I\, Z\, m_p)$. The exact relativistic result for the electric-quadrupole correction is [13]

$$\delta\sigma_Q = -\frac{\alpha\,(Z\alpha)^3\,Q\,m}{I(2I-1)\,g_I\,m_p}\,\frac{6\left[35 + 20\gamma - 32(Z\alpha)^2\right]}{45\,\gamma(1+\gamma)^2\left[15 - 16(Z\alpha)^2\right]}\,, \quad (5)$$

where Q is the nuclear electric quadrupole moment and $\gamma = \sqrt{1-(Z\alpha)^2}$.

We now turn to the QED correction to the nuclear magnetic shielding. It consists of the self-energy (SE) and vacuum-polarization parts, the SE being the most difficult one. The Feynman diagrams representing the SE correction (Fig. 1) contain two magnetic interactions, one with the external magnetic field (in what follows, the Zeeman interaction), $V_{\rm zee}(r) = \frac{|e|}{2}\boldsymbol{B}\cdot(\vec{r}\times\vec{\alpha})$, and the other with the magnetic dipole nuclear field (in what follows, the hfs interaction), $V_{\rm hfs}(r) = \frac{|e|}{4\pi}\boldsymbol{\mu}\cdot(\vec{r}\times\vec{\alpha})/r^3$. Formal expressions for the corresponding energy shifts can be obtained by the two-time Green's function method [15]. Irreducible parts of the diagrams in Fig. 1(a)-(c) give rise to the *perturbed orbital* contribution,

$$\delta E_{\rm po} = 2\,\langle a|\Sigma(\varepsilon_a)|\delta^{(2)}a\rangle + 2\,\langle \delta^{(1)}_{\rm hfs}a|\Sigma(\varepsilon_a)|\delta^{(1)}_{\rm zee}a\rangle\,, \quad (6)$$

where Σ is the SE operator, $|\delta^{(1)}_{\rm zee}a\rangle$ and $|\delta^{(1)}_{\rm hfs}a\rangle$ are the first-order perturbations of the reference-state wave function induced by $V_{\rm zee}$ and $V_{\rm hfs}$, respectively, and $|\delta^{(2)}a\rangle$ is the second-order perturbation induced by both interactions. The SE operator is defined by

$$\langle i|\Sigma(\varepsilon)|k\rangle = \frac{i}{2\pi}\int_{-\infty}^{\infty} d\omega \sum_n \frac{\langle in|I(\omega)|nk\rangle}{\varepsilon - \omega - u\varepsilon_n}\,, \quad (7)$$

where $I(\omega) = e^2\alpha_\mu\alpha_\nu D^{\mu\nu}(\omega)$, $D^{\mu\nu}(\omega)$ is the photon propagator, and $u \equiv 1 - i0$. The diagram in Fig. 1(d) gives rise to the *hfs-vertex* contribution,

$$\delta E_{\rm vr,hfs} = 2\,\langle a|\Gamma_{\rm hfs}(\varepsilon_a)|\delta^{(1)}_{\rm zee}a\rangle + 2\,\langle a|\Sigma'|\delta^{(1)}_{\rm zee}a\rangle\,\langle V_{\rm hfs}\rangle\,, \quad (8)$$

where the prime denotes the derivative of the operator with respect to the energy argument and

$$\langle i|\Gamma_{\rm hfs}(\varepsilon)|k\rangle = \frac{i}{2\pi}\int_{-\infty}^{\infty} d\omega \times \sum_{n_1 n_2}\frac{\langle in_2|I(\omega)|n_1k\rangle\langle n_1|V_{\rm hfs}|n_2\rangle}{(\varepsilon-\omega-u\varepsilon_{n_1})(\varepsilon-\omega-u\varepsilon_{n_2})}\,. \quad (9)$$

The diagram in Fig. 1(e) induces the *Zeeman-vertex* contribution, in analogy with its hfs-vertex counterpart,

$$\delta E_{\rm vr,zee} = 2\,\langle a|\Gamma_{\rm zee}|\delta^{(1)}_{\rm hfs}a\rangle + 2\,\langle a|\Sigma'|\delta^{(1)}_{\rm hfs}a\rangle\,\langle V_{\rm zee}\rangle. \quad (10)$$

Finally, Fig. 1(f) together with the remaining derivative terms yields the *double-vertex* contribution,

$$\delta E_{\rm d.vr} = 2\,\langle\Lambda\rangle + \langle\Sigma''\rangle\langle V_{\rm zee}\rangle\langle V_{\rm hfs}\rangle + \langle\Gamma'_{\rm hfs}\rangle\langle V_{\rm zee}\rangle + \langle\Gamma'_{\rm zee}\rangle\langle V_{\rm hfs}\rangle + 2\,\langle\Sigma'\rangle\,\langle a|V_{\rm zee}|\delta^{(1)}_{\rm hfs}a\rangle, \quad (11)$$

TABLE I: QED corrections to the nuclear magnetic shielding.

Z	SE	VP
10	$-0.51\,(10)$	0.229
14	$-0.710\,(15)$	0.256
16	$-0.789\,(9)$	0.271
20	$-0.927\,(4)$	0.302
26	$-1.110\,(2)$	0.355
32	$-1.283\,(1)$	0.417
40	$-1.519\,(1)$	0.520
54	$-2.029\,(1)$	0.775
82	$-4.457\,(2)$	1.996
92	$-7.107\,(2)$	2.954

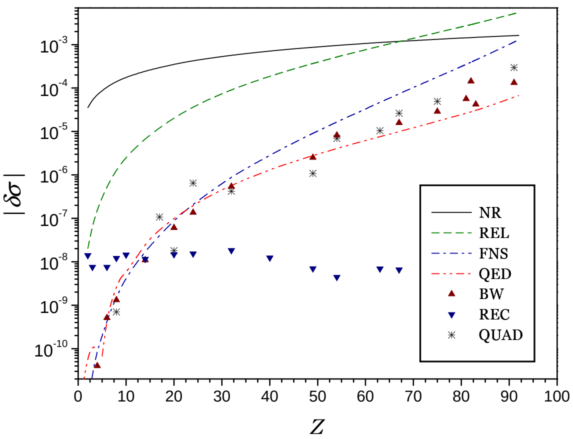

FIG. 2: (Color online) Individual contributions to the nuclear shielding. "NR" is the nonrelativistic contribution, "REL" is the relativistic point-nucleus contribution, "FNS" is the finite nuclear size correction, "QED" is the QED correction, "BW" is the Bohr-Weisskopf correction, "REC" is the recoil correction, and "QUAD" is the electric quadrupole correction. Note that the QED correction changes its sign between $Z = 4$ and 5.

where $\Lambda \equiv \Lambda(\varepsilon_a)$ is the 4-point vertex operator,

$$\langle i|\Lambda(\varepsilon)|k\rangle = \frac{i}{2\pi}\int_{-\infty}^{\infty} d\omega \sum_{n_1 n_2 n_3} \times \frac{\langle i n_3|I(\omega)|n_1 k\rangle\langle n_1|V_{\rm Zee}|n_2\rangle\langle n_2|V_{\rm hfs}|n_3\rangle}{(\varepsilon-\omega-u\varepsilon_{n_1})(\varepsilon-\omega-u\varepsilon_{n_2})(\varepsilon-\omega-u\varepsilon_{n_3})}\,. \tag{12}$$

The formulas reported so far refer to the energy shifts. The corrections to the magnetic shielding are related to the energy shifts by $\delta\sigma_i = \delta E_i\, I F(F+1)/(\mu\, B\, M_F\, \langle \boldsymbol{I}\cdot\boldsymbol{F}\rangle)$, where M_F is the projection of the total momentum F. It can be shown that for the $j = 1/2$ reference states, $\delta\sigma_{\rm QED}$ does not depend on nuclear quantum numbers. The numerical calculation of $\delta\sigma_{\rm SE}$ was performed along the lines developed in Ref. [16]; its details will be reported elsewhere.

The remaining part of the QED effect is the vacuum polarization (VP). In our calculation, we include two dominant VP corrections induced by (i) modification of the electron line by the Uehling potential and (ii) modification of the hfs interaction by the free-loop VP.

Our calculational results for the SE and VP corrections are listed in Table I, expressed in terms of the function $D(Z\alpha)$,

$$\delta\sigma_{\rm QED} = \alpha^2\,(Z\alpha)^3\,D(Z\alpha)\,. \tag{13}$$

The SE correction is calculated for the point nucleus, whereas the VP part accounts for the finite nuclear size as well as higher-order iterations of the Uehling potential. Because of large numerical cancellations, we were able to perform our numerical SE calculations for $Z \geq 10$ only. In order to extend our calculations to the lower-Z ions and to cross-check the numerical procedure, we also performed an analytical calculation of the leading term of the $Z\alpha$ expansion. The result valid for an ns state reads

$$D_n(Z\alpha) = \frac{8}{9\pi n^3}\left[\ln(Z\alpha)^{-2} + 2\,\ln k_0 - 3\,\ln k_3 - \frac{1817}{480}\right], \tag{14}$$

where $\ln k_0(1s) = 2.984\,128$ and $\ln k_3(1s) = 3.272\,806$ [9]. Details of the analytical calculation will be reported elsewhere. The results of the numerical and the analytical calculations are in good agreement.

We now turn to the effect induced by the spatial distribution of the nuclear magnetic moment, also known as the Bohr-Weisskopf (BW) correction. Following Ref. [17], our treatment of the BW effect is based on the effective single-particle model of the nuclear magnetic moment. Within this model, the magnetic moment is assumed to be induced by the odd nucleon with an effective g-factor, which is fitted to yield the experimental value of the nuclear magnetic moment. Under these assumptions, the BW effect can be described by the magnetization-distribution function $F(r)$ that multiplies the standard point-dipole hfs interaction $V_{\rm hfs}(r)$. The function $F(r)$ is induced by the wave function of the odd nucleon, which is obtained by solving the Schrödinger equation with the Woods-Saxon potential (see Ref. [18] for details). The BW correction $\delta\sigma_{\rm BW}$ is obtained by reevaluating the leading-order magnetic shielding $\sigma^{(0)}$ with the hfs interaction $V_{\rm hfs}$ multiplied by the magnetization-distribution function $F(r)$. The relative uncertainty of 30% is ascribed to this correction, which is consistent with previous error estimates for this effect [17].

Numerical results of our calculations are presented in Table II and Fig. 2. The error of the QED correction comes from the numerical uncertainty of the SE part and the estimate of uncalculated VP terms (30% of the total VP part). The error of the quadrupole contribution comes from the nuclear quadrupole moments. The largest error is due to the BW correction. Since this effect cannot be presently accurately calculated, this uncertainty sets the practical limit to which the nuclear magnetic moment can be determined from an atomic system.

TABLE II: Individual contributions to the shielding constant $\sigma \times 10^6$ for selected hydrogen-like ions, see Eq. (3).

	$^{17}O^{7+}$	$^{43}Ca^{19+}$	$^{73}Ge^{31+}$	$^{131}Xe^{53+}$	$^{209}Bi^{82+}$
Leading	143.3127	375.960	657.93	1461.6	4112
QED	−0.0026 (2)	−0.103 (15)	−0.59 (8)	−4.1 (0.8)	−30 (7)
Bohr-Weisskopf	−0.0013 (4)	−0.061 (18)	−0.54 (16)	−8.2 (2.5)	−42 (13)
Quadrupole	−0.0007 (1)	−0.018	−0.42	6.9 (0.1)	7
Recoil	−0.0120	−0.015	−0.02	0.0	0
Total	143.2960 (5)	375.763 (24)	656.36 (18)	1456.3 (2.6)	4046 (15)

For very light ions, the theoretical accuracy is limited by the recoil effect (see Fig. 2), which is known in the nonrelativistic limit only. Note that some of corrections to σ depend on the nuclear g-factor. This dependence, however, is so weak that it can be safely ignored in the determination of the magnetic moments.

Summarising, we have presented *ab initio* calculations of the nuclear shielding in hydrogen-like ions, which account for relativistic, nuclear, and QED effects. The present theory permits determination of nuclear magnetic moments with fractional accuracy ranging from 10^{-9} in the case of $^{17}O^{7+}$ to 10^{-5} for $^{209}Bi^{82+}$. This Letter is primarily focused on nuclei with spin $I > 1/2$, but the case of $I = 1/2$ is only slightly more complicated. Then, the nuclear-spin-independent part of g_F in Eq. (1) can be cancelled approximately, by taking a difference of the g-factors g_F for two different isotopes of the same element.

Modern experiments on g-factors of hydrogen-like ions have achieved the accuracy of a few parts in 10^{11} [19] but so far have been restricted to ions with spinless nuclei. Their extention to the nuclei with spin requires driving the hfs transition and measuring the g-factor of an atom in a hyperfine excited state. These are significant complications but they do not make an experiment prohibitively difficult [19].

Stimulating discussions with K. Blaum and G. Werth are gratefully acknowledged. Z.H. was supported by the Alliance Program of the Helmholtz Association (HA216/EMMI). V.A.Y. was supported by the Helmholtz Association (Nachwuchsgruppe VH-NG-421). K.P. acknowledges support by NIST Precision Measurement Grant PMG 60NANB7D6153.

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