
Relativistic Perturbation Theory Calculation of the Hyperfine Structure Parameters for Some Heavy-Element Isotopes

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ABSTRACT: The energies and hyperfine structure constants for some Li-like multicharged ions and atoms of ^{133}Cs and ^{181}Hg are calculated within the quantum electrodynamics (QED) perturbation theory formalism with correct taking into account the correlation, relativistic, nuclear, and radiative corrections. © 2009 Wiley Periodicals, Inc. *Int J Quantum Chem* 109: 3330–3335, 2009

Key words: hyperfine structure; QED theory; correlation; nuclear; radiative corrections

1. Introduction

At present time, studying the spectral lines hyperfine structure for heavy elements and multicharged ions is of a great interest for the further development as atomic and nuclear theories and spectroscopy of multicharged ions (see, for example, Refs. [1–22]). One could also mention here the important astrophysical applications. The experiments on the definition of hyperfine splitting also enable to refine the deduction of nuclear magnetic moments of different isotopes and to check an ac-

curacy of the various calculational models used for the theoretical description of the nuclear effects. The multiconfiguration relativistic Hartree–Fock (RHF) and Dirac–Fock (DF) approaches (see, for example, Refs. [3–5, 8–18]) are the most reliable versions of calculation for multielectron systems with a large nuclear charge. Usually, in these calculations the one- and two-body relativistic effects are taken into account practically precisely. It should be given the special attention to three very general and important computer systems for relativistic and QED calculations of atomic and molecular properties developed in the Oxford and German-Russian groups etc. (“GRASP,” “Dirac,” “BERTHA,” “QED,” “Dirac”) (see Refs. [3–5, 8–18] and references therein). For example, the BERTHA

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program embodies a new formulation of relativistic atomic and molecular structure theory within the framework of relativistic QED. This leads to a simple and transparent formulation of the Dirac–Hartree–Fock–Breit self-consistent field equations along with algorithms for molecular properties, electron correlation, and higher order QED effects. These equations are solved by a direct method based on a relativistic generalization of the McMurchie–Davidson algorithm for corresponding integrals that economizes memory requirements and is not significantly more expensive computationally than comparable nonrelativistic calculations. The useful overview of the relativistic electronic structure theory is presented in Refs. [12, 13, 17–20] from the QED point of view. The next important step is an adequate taking into account the QED corrections. This topic has been a subject of intensive theoretical and experimental interest (see, for example [3–5, 8–20]).

In this article, an effective ab initio QED approach [11, 14, 20–23] to calculation of the spectra for multielectron heavy ions with taking into account the relativistic, correlation, nuclear, radiative effects is applied to the relativistic calculation of the hyperfine structure parameters for the Li-like multicharged ions and cesium and mercury atoms. The calculation scheme is based on the gauge-invariant QED perturbation theory (PT) with using the optimized one-quasiparticle representation in the theory of relativistic systems [11]. There have taken into account all correlation corrections of the second order and dominated classes of the higher orders diagrams (electrons screening, mass operator iterations etc.) [11, 21, 22]. The magnetic interelectron interaction is accounted for in the lowest order on α^2 (α is the fine structure constant) parameter. The Lamb shift polarization part is taken into account in the modified Uehling–Serber approximation. The Lamb shift self-energy part is accounted for effectively within the generalized Ivanov–Ivanova nonperturbative procedure [11].

2. Theory

Let us describe the key moments of the approach (more details can be found in Refs. [11, 14, 20–23]). The electron wave functions (the PT zeroth basis) are found from solution of the relativistic Dirac equation with potential, which includes ab initio mean-field potential, electric, polarization potentials of a nucleus. The charge distribution in the

Li-like ion and the mercury nuclei is modeled within the Gauss model. The nuclear model used for the Cs isotope is the independent particle model with the Woods–Saxon and spin-orbit potentials (see Ref. [24]). Although we have no guaranty that these wave-functions yield a close approximation to nature, its acceptability is checked in calculating the metastable discharge of a nucleus by capture of the negative muon [24].

Let us consider in details more simple case of the Li-like ion. We set the charge distribution in the Li-like ion nucleus $\rho(r)$ by the Gaussian function. With regard to normalization we have:

$$\rho(r|R) = (4\gamma^{3/2}/\sqrt{\pi})\exp(-\gamma r^2) \quad (1)$$

where $\gamma = 4/\pi R^2$ and R is the effective nucleus radius. The Coulomb potential for the spherically symmetric density $\rho(r)$ is:

$$V_{\text{nuc}}(r|R) = -((1/r)\int_0^r dr' r'^2 \rho(r'|R) + \int_r^\infty dr' r' \rho(r'|R)) \quad (2)$$

It is determined by the following system of differential equations:

$$V_{\text{nuc}}'(r, R) = (1/r^2)\int_0^r dr' r'^2 \rho(r', R) \equiv (1/r^2)y(r, R) \quad (3)$$

$$y'(r, R) = r^2 \rho(r, R) \quad (4)$$

$$\begin{aligned} \rho'(r, R) &= -8\gamma^{5/2}r/\sqrt{\pi}\exp(-\gamma r^2) = -2\gamma r \rho(r, R) \\ &= -\frac{8r}{\pi r^2} \rho(r, R) \end{aligned} \quad (5)$$

with the corresponding boundary conditions. Consider the DF type equations for a three-electron system $1s^2nlj$. Formally, they fall into one-electron Dirac equations for the orbitals $1s$ and nlj with the potential:

$$V(r) = 2V(r|1s) + V(r|nlj) + V_{\text{ex}}(r) + V(r|R) \quad (6)$$

$V(r, R)$ includes the electrical and the polarization potentials of the nucleus; the components of the Hartree potential (in the Coulomb units):

$$V(r|i) = \frac{1}{Z} \int d\vec{r}' \rho(r|i) / |\vec{r} - \vec{r}'| \quad (7)$$

Here $\rho(r|i)$ is the distribution of the electron density in the state i , V_{ex} is the exchange interelectron interaction. The main exchange effect will be taken into account if in the equation for the valent electron orbital we assume $V(r) = V(r|\text{core}) + V(r|nlj)$ and in the equation for the nlj orbital $V(r) = 2V(r|\text{core})$. The rest of the exchange and correlation effects will be taken into account in the first two orders of the PT by the total interelectron interaction [11, 14, 21, 22].

A procedure of taking into account the radiative QED corrections is in details given in the Refs. [11, 14, 20–22]. Regarding the vacuum polarization effect, let us note that this effect is usually taken into consideration in the first PT theory order by means of the Uehling potential. This potential is usually written as follows:

$$U(r) = -\frac{2\alpha}{3\pi r} \int_1^\infty dt \exp(-2rt/\alpha Z) \left(1 + 1/2t^2 \right) \frac{\sqrt{t^2 - 1}}{t^2} \equiv -\frac{2\alpha}{3\pi r} C(g), \quad (8)$$

where $g = r/(\alpha Z)$. In our calculation, we use more exact approach [22]. The Uehling potential, determined as a quadrature (8), may be approximated with high precision by a simple analytical function. The use of new approximation of the Uehling potential permits one to decrease the calculation errors for this term down to 0.5–1%. A method for calculation of the self-energy part of the Lamb shift is based on an idea by Ivanov-Ivanova (see Ref. [11]). In an atomic system the radiative shift and the relativistic part of energy are, in principle, defined by one and the same physical field. It may be supposed that there exists some universal function that connects the self-energy correction and the relativistic energy. The self-energy correction for the states of a hydrogen-like ion was presented by Mohr [9] as:

$$E_{\text{SE}}(H|Z, nlj) = 0.027148 \frac{Z^4}{n^3} F(H|Z, nlj) \quad (9)$$

These results are modified here for the states $1s^2 nlj$ of Li-like ions. It is supposed that for any ion with

nlj electron over the core of closed shells the sought value may be presented in the form:

$$E_{\text{SE}}(Z, nlj) = 0.027148 \frac{\xi^4}{n^3} f(\xi, nlj) (\text{cm}^{-1}) \quad (10)$$

The parameter $\xi = (E_{\text{R}})^{1/4}$, E_{R} is the relativistic part of the bounding energy of the outer electron; the universal function $f(\xi, nlj)$ does not depend on the composition of the closed shells and the actual potential of the nucleus. The procedure of generalization for a case of Li-like ions with a finite nucleus consists of the following steps: (i) calculation of the values E_{R} and ξ for the states nlj of H-like ions with the point nucleus (in accordance with the Sommerfeld formula); (ii) construction of an approximating function $f(\xi, nlj)$ by the found reference Z and the appropriate $F(H|Z, nlj)$; (iii) calculation of E_{R} and ξ for the states nlj of Li-like ions with a finite nucleus; (iv) calculation of E_{SE} for the sought states by the formula [10].

The energies of the states of Li-like ions were calculated twice: with a conventional constant of the fine structure $\alpha = 1/137.04$ and with $\tilde{\alpha} = \alpha/1,000$. The results of latter calculations were considered as nonrelativistic. This permitted isolation of E_{R} and ξ . A detailed evaluation of their accuracy may be made only after a complete calculation of $E_{\text{SE}}^{\text{nr}}(\text{Li } Z, nlj)$. It may be stated that the above extrapolation method is more justified than using the widely spread expansions by the parameter αZ .

The energies of electric quadruple and magnetic dipole interactions are defined by a standard way with the hyperfine structure constants, usually expressed through the standard radial integrals [25]:

$$A = \{[(4,32587)10^{-4}Z_{\text{Xg1}}^2]/(4\chi^2 - 1)\}(RA)_{-2}, \quad (11a)$$

$$B = \{7.2878 \cdot 10^{-7}Z^3Q/[(4\chi^2 - 1)I(I - 1)]\}(RA)_{-3}, \quad (11b)$$

Here g_I is the Lande factor, Q is a quadruple momentum of nucleus (in Barn); $(RA)_{-2}$, $(RA)_{-3}$ are the radial integrals usually defined as follows:

$$(RA)_{-2} = \int_0^\infty dr r^2 F(r) G(r) U(1/r^2, R), \quad (12a)$$

TABLE I
The hyperfine structure constants of some Li-like ions: $A = Z^3 g_I \bar{A}$ (cm⁻¹) and $B = Z^3 Q_B / [I(2I-1)]$ (cm⁻¹).

| nlj | Z | 20 | 69 | 79 | 92 |
|-------------------|-----------|-------|-------|-------|--------|
| 3s | \bar{A} | 26-03 | 51-03 | 63-03 | 90-03 |
| 4s | \bar{A} | 15-03 | 19-03 | 24-03 | 36-03 |
| 2p _{1/2} | \bar{A} | 25-03 | 56-03 | 71-03 | 105-02 |
| 3p _{1/2} | \bar{A} | 81-04 | 16-03 | 20-03 | 31-03 |
| 4p _{1/2} | \bar{A} | 32-04 | 72-04 | 91-04 | 11-03 |
| 2p _{3/2} | \bar{A} | 50-04 | 67-04 | 71-04 | 72-04 |
| | \bar{B} | 9-04 | 13-04 | 15-04 | 17-04 |
| 3p _{3/2} | \bar{A} | 13-04 | 19-04 | 21-04 | 22-04 |
| | \bar{B} | 31-05 | 51-05 | 55-05 | 62-05 |
| 4p _{3/2} | \bar{A} | 62-05 | 89-05 | 92-05 | 8-04 |
| | \bar{B} | 10-05 | 20-05 | 22-05 | 26-05 |
| 3d _{3/2} | \bar{A} | 88-05 | 10-04 | 11-04 | 12-04 |
| | \bar{B} | 51-06 | 9-05 | 10-05 | 11-05 |
| 4d _{3/2} | \bar{A} | 35-05 | 51-05 | 55-05 | 58-05 |
| | \bar{B} | 12-06 | 44-06 | 50-06 | 56-06 |
| 3d _{5/2} | \bar{A} | 36-05 | 48-05 | 50-05 | 52-05 |
| | \bar{B} | 21-06 | 38-06 | 39-06 | 40-06 |
| 4d _{5/2} | \bar{A} | 15-05 | 19-05 | 20-05 | 21-05 |
| | \bar{B} | 59-07 | 15-06 | 16-06 | 17-06 |
| 4f _{5/2} | \bar{A} | 06-05 | 12-05 | 13-05 | 14-05 |
| | \bar{B} | 16-07 | 53-07 | 58-07 | 63-07 |
| 4f _{7/2} | \bar{A} | 61-06 | 78-06 | 81-06 | 83-06 |

$$(RA)_{-3r} = \int_0^\infty dr r^2 [F^2(r) + G^2(r)] U(1/r^2, R). \quad (12b)$$

The radial parts F and G of the Dirac function two components for electron, which moves in the potential $V(r, R) + U(r, R)$, are determined by solution of the Dirac equations. To define the hyperfine interaction potentials $U(1/r^n, R)$, we use the method by Ivanov et al. [11]. Other details of the used method can be found in the references [11, 14, 21–24].

3. Results of Calculating the Hyperfine Structure Parameters and Discussion

In Table I, we present the calculated data of the hyperfine structure constants for some Li-like ions. There are presented results for the following parameters: $A = Z^3 g_I \bar{A}$ and $B = Z^3 Q / [I(2I - 1)] \bar{B}$.

In Table II, the experimental (A^{Exp}) and theoretical data of the magnetic dipole constant A (MHz) for the valent states of ¹³³Cs atom ($I = 7/2$, $g_I = 0.7377208$) are presented. The theoretical results are obtained on the basis of the standard RHF (A^{RHF}) calculation, the RHF ($A^{\text{RHF}} + dA$) calculation with taking into account the PT second and higher corrections (see Ref. [15] and references therein) and the QED PT (A^{QED}) calculation (this work). The analysis shows that taking into account the correlation and QED corrections is important to reach the physically reasonable agreement between theoretical and experimental data.

Further we present the experimental data and theoretical results (see Table III) for the energies, hyperfine structure constants, and electric quadrupole nuclear moment Q for the isotope of ¹⁸¹Hg. This atom has the external valent shell $6s^2$ and can be naturally treated as the two-quasiparticle system. The corresponding formula (11), (12) are generalized on this case by standard way [25]. In this Table III, we present the results of calculation of the hyperfine structure constants (in MHz) for ³P₁ state of mercury on the basis of different methods, namely: standard uncorrelated DF approach, multiconfiguration DF (MCDF) method with taking into account the Breit and QED corrections, and the QED PT approach (see Refs. [8, 21]).

In Table IV, we present the values of the electric quadrupole moment Q (in mBarn) for the isotope of ¹⁸¹Hg. Different experimental methods have been used and listed in the table. The corresponding theoretical results of calculation within the MCDF method (with taking into account the Breit and

TABLE II
The values (in MHz) of the hyperfine structure constant A for valent states of the ¹³³Cs isotope: A^{Exp} , experiment; A^{RHF} , the RHF calculation data; $A^{\text{RHF}} + dA^{\text{RHF}}$, the RHF calculation data with taking into account the PT second and higher orders contributions [15]; A^{QED} , the QEDPT calculation data (this work).

| State | A^{RHF} | $A^{\text{RHF}} + dA$ | A^{QED} | A^{Exp} |
|-------------------|------------------|-----------------------|------------------|------------------|
| 6s _{1/2} | 1426.81 | 2291.00 | 2294.45 | 2298.16 |
| 7s _{1/2} | 392.05 | 544.04 | 545.480 | 545.90(9) |
| 6p _{1/2} | 161.09 | 292.67 | 292.102 | 291.90(13) |
| 7p _{1/2} | 57.68 | 94.21 | 94.317 | 94.35(4) |
| 6p _{3/2} | 23.944 | 49.785 | 50.205 | 50.275(3) |
| 7p _{3/2} | 8.650 | 16.255 | 16.590 | 16.605(6) |
| 3d _{3/2} | — | — | 16.422 | — |

TABLE III

The experimental and calculated values of the nuclear quadrupole moment Q (mB) and hyperfine structure constants (MHz) for the $3p$, state of the neutral mercury ^{201}Hg .

| Calculation method | Q (mb) | A (MHz) | B (MHz) |
|--------------------|--------------|-------------------|------------------|
| DF | 478.13 | -4368.266 | |
| MCDF (+Breit+QED) | 386.626 | -5470.810 | |
| QED PT | 380.518 | -5460.324 | -286.512 |
| Experiment | Look table 4 | -5454.569 (0.003) | -280.107 (0.005) |

QED corrections) and QED PT method are presented too. The detailed analysis shows that our value for the moment Q is best of all agreed with the result by Ulm et al.

The fundamental reason of physically reasonable agreement between theory and experiment is connected with the correct taking into account the interelectron correlation effects, nuclear (due to the finite size of a nucleus), relativistic, and radiative corrections. The key difference between the results of the RHF, MCDF, QED PT methods calculations is explained by using the different schemes of taking into consideration the interelectron correlations. The contribution of the PT high order effects and nuclear contribution may reach the units and even

dozens of MHz and should be correctly taken into account. It is necessary to take into account more correctly the spatial distribution of the magnetic moment inside a nucleus (the Bohr-Weisskopf effect), the nuclear-polarization corrections, etc. too. One could note that the simple single-particle nuclear models are usually (see, for example, Refs. [26]) used for the evaluation of the Bohr-Weisskopf correction. It can be done within nuclear modeling with using the shell model with the Woods-Saxon and spin-orbit potentials or relativistic mean field approach [3, 6, 24, 26]. In last years, an account for the Bohr-Weisskopf effect and the high-order QED contributions is considered in many papers (see, for example, [3-7, 16, 24, 26, 27]). In any case, it is obvious that the contributions due to these effects and nonaccount of the QED high-order corrections are in part responsible for difference between presented theoretical results and experimental data.

TABLE IV

The values of electric quadrupole moment q (in mBarn) for isotope of ^{201}Hg .

| Q (B) | Method | Reference | Year |
|------------|------------------------------------|-----------------------|------|
| 380,5 | Atomic | This paper | 2008 |
| 387 (6) | Atomic | Pyykko et al | 2005 |
| 347 (43,0) | Nuclear | Fornal et al | 2001 |
| 385 (40) | Atomic ^a | Ulm et al | 1988 |
| 485 (68) | Muonic ^b | Gunther et al | 1983 |
| 386 (49) | Muonic $3d^c$ | Hahn et al | 1979 |
| 267 (37) | Muonic $2p^c$ | Hahn et al | 1979 |
| 390 (20) | Solid ^d | Edelstein and Pound | 1975 |
| 455 (40) | Atomic ³ P ₂ | McDermott and Lichten | 1960 |
| 420 | Atomic | Murakawa | 1959 |
| 500 (50) | Atomic | Blaise and Chantrel | 1957 |
| 600 | Solid | Dehmelt et al | 1954 |
| 500 | Atomic | Schuler and Schmidt | 1935 |

^a Standard value by Raghavan.

^b Value for ^{199}Hg ($l = 5/2$), which is agreed with relation 201/199.

^c Direct muon experiment for ^{201}Hg .

^d Solid value for HgCl_2 plus compiled value for ^{199}Hg (see [8a]).

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