Isotope shifts of the 1s²2s²2p(J) − 1s²2s² transition energies in Be-like thorium and uranium

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Abstract

Precise calculations of the isotope shifts in beryllium-like thorium and uranium ions are presented. The main contributions to the field and mass shifts are calculated within the framework of the Dirac–Coulomb–Breit Hamiltonian employing the configuration-interaction Dirac–Fock–Sturm method. These calculations include the relativistic, electron–electron correlation, and Breit-interaction effects. The QED, nuclear deformation, and nuclear polarization corrections are also evaluated.

Keywords: highly charged ions, isotope shifts, nuclear charge radius

1. Introduction

Theoretical and experimental studies of the isotope shifts in highly charged ions can provide tests of the relativistic and QED theory of the nuclear recoil effect in nonperturbative regime and serve as a good tool for precise determination of the nuclear charge radius differences. First isotope shift measurements in highly charged ions were performed in [1–3]. The measurements of the isotope shifts of the binding energies in B-like argon [4] and in Li-like neodymium [5] have provided the first tests of the relativistic theory of the nuclear recoil effect with highly charged ions. The latter experiment led also to determination of the nuclear charge radius difference for the ¹⁴²,¹⁵²Nd isotopes. The use of the dielectronic recombination technique [5–7] at the GSI Helmholtzzentrum für Schwerionenforschung (GSI) [8] and the Facility for Antiproton and Ion Research (FAIR) [9] allows the related experiments for heavy Be-like ions.

The main goal of the present work is to extend our calculations of the isotope shifts in Li- and B-like ions [10, 11] to Be-like thorium and uranium ions, which are of most interest for the experimental study. Previously, the calculations of the isotope shifts in Be-like ions have been performed in the range Z = 5 − 74 with the use of the multiconfiguration Dirac–Fock (DF) method [12]. The calculations of the transition energies in these ions have been performed in [13, 14] (see also [15, 16] and references therein).

The precision of the isotope shift measurements in heavy ions is approaching the level of the QED effects. Moreover, it is expected that at the FAIR facilities this precision will be
improved by an order of magnitude. It means that the relevant theoretical calculations must be performed to the utmost accuracy. In the present paper, the dominant contributions to the isotope shifts are calculated employing the Dirac–Coulomb–Breit Hamiltonian. These calculations, which are based on the Dirac–Fock–Sturm method [17, 18], include the relativistic, electron–electron correlation, and Breit-interaction effects. Additionally, we evaluate the QED, nuclear deformation, and nuclear polarization corrections which become rather large for heavy ions.

The relativistic units \((\hbar = c = 1)\) are used throughout the paper.

2. Theory

2.1. Method of calculation

In the present work, the large-scale configuration-interaction DF method was used to solve the Dirac–Coulomb–Breit equation and to calculate the isotope shifts in Be-like uranium and thorium. The many-electron wave function \(\psi(\gamma J)\) with quantum numbers \(\gamma\) and \(J\) was expanded in terms of a large number of the configuration state functions (CSFs) with the same \(J\):

\[
\psi(\gamma J) = \sum_{\alpha} c_{\alpha} \Phi_{\alpha}(J),
\]

(1)

For every relativistic atomic configuration the CSFs \(\Phi_{\alpha}(J)\) are eigenfunctions of the square of total angular momentum \(J^2\). They are obtained as the linear combinations of the Slater determinants, which have been constructed employing the Dirac–Fock–Sturm method [17, 18]. In this approach, the orbitals corresponding to the occupied shells are obtained by the DF method, while the other vacant orbitals are determined by solving the Dirac–Fock–Sturm equations

\[
(h^{\text{DF}} - \varepsilon_{\alpha}) \phi_{\alpha} = \lambda_{f} W(r) \phi_{f},
\]

(2)

where \(h^{\text{DF}}\) is the DF operator, \(\varepsilon_{\alpha}\) is the one-electron energy of the occupied DF orbital, and \(W(r)\) is a constant sign weight function. The parameter \(\lambda_{f}\) plays a role of an eigenvalue of the Sturmian operator. The weight function \(W(r)\) is chosen to be regular at the origin and goes to zero like \(1/r^2\) as \(r \to \infty\). As a result, the Sturmian eigenfunctions form a discrete and complete basis set of one-electron wave functions.

2.2. Nuclear size effect

The finite nuclear size effect (the so-called field shift) is caused by the difference in the nuclear charge distribution of the isotopes. The main contribution to the field shift can be calculated in the framework of the Dirac–Coulomb–Breit Hamiltonian. The nuclear charge distribution is usually approximated by the spherically-symmetric Fermi model

\[
\rho(r, R) = \frac{N}{1 + \exp[(r - c)/a]},
\]

(3)

where the parameter \(a\) is generally fixed to be \(a = 2.3/(4 \ln 3)\) fm and the parameters \(N\) and \(c\) are determined using the given value of the root-mean-square (rms) nuclear charge radius \(R = \langle r_r^2 \rangle^{1/2}\) and the normalization condition \(\int d\rho(r, R) = 1\). The potential induced by the nuclear charge distribution \(\rho(r, R)\) is defined as

\[
V_N(r, R) = -4\pi \alpha Z \int_0^\infty dr' r'^2 \rho(r', R) \frac{1}{r_s^2},
\]

(4)

where \(r_s = \max(r, r')\). Since the finite nuclear size effect is mainly determined by the rms nuclear charge radius, the energy difference between two isotopes can be approximated as

\[
\delta E_{FS} = F \delta \langle r^2 \rangle,
\]

(5)

where \(F\) is the field shift factor and \(\delta \langle r^2 \rangle\) is the mean-square charge radius difference. In accordance with this definition and the virial theorem, the \(F\) factor can be also evaluated by

\[
F = \langle \psi | \sum_i dV_N(r_i, R) d\langle r_i^2 \rangle | \psi \rangle,
\]

(6)

where \(\psi\) is the wave function of the state under consideration and the index \(i\) runs over all atomic electrons.

2.3. Relativistic nuclear recoil effect

The fully relativistic theory of the nuclear recoil effect can be formulated only in the framework of quantum electrodynamics [19–23]. However, to the lowest relativistic order (within the Breit approximation), the nuclear recoil effect can be taken into account using the effective recoil operator [19, 20, 24]:

\[
H_{\text{rel}} = \frac{1}{2M} \sum_{i,k} \left[ p_i \cdot p_k - \frac{\alpha Z}{r_i} \left[ \alpha_i + \frac{(\alpha_i \cdot r_i) r_i}{r_i^2} \right] \cdot p_k \right].
\]

(7)

This operator can be used for relativistic calculations of the nuclear recoil effect in ions and atoms (see, e.g. [10–12, 17, 18, 25, 26] and references therein). The calculation is carried out by averaging the operator (7) with the eigenvectors of the Dirac–Coulomb–Breit Hamiltonian.

2.4. QED, nuclear deformation and nuclear polarization corrections

Since our consideration is restricted to very heavy ions, the independent-electron approximation can be used to evaluate the QED, nuclear deformation, and nuclear polarization corrections.

To calculate the self-energy and vacuum-polarization corrections to the field shift, one can use analytical formulas for these corrections derived for H-like ions in [27]. In case of uranium, an approximate formula obtained by fitting the direct numerical calculations [28] can be also employed.

The QED calculation of the one-electron recoil effect for \(n = 1, 2\) states was performed in [23, 29–31]. In addition, two-electron recoil contributions of zeroth order in \(1/Z\) should be taken into account for the He-like ions (1s2s2p(J)) states. A detailed analysis of the relevant contributions for He-like ions was presented in [31].
Table 1. Individual contributions to the isotope shifts of the $1s^22s2p_{1/2}(J) - 1s^22s^2$ energy differences in $^{232,230}\text{Th}^{86+}$, $^{238,236}\text{U}^{88+}$, and $^{238,234}\text{U}^{88+}$ (in meV) with given values of $\delta \langle r^2 \rangle$.

|                         | $^{232,230}\text{Th}^{86+}$ | $^{238,236}\text{U}^{88+}$ | $^{238,234}\text{U}^{88+}$ |
|-------------------------|-----------------------------|-----------------------------|-----------------------------|
| Main contributions      |                             |                             |                             |
| Field shift             | -112.4                      | -110.8                      | -220.7                      |
| Mass shift              | 0.1                         | 0.1                         | 0.2                         |
| QED                     |                             |                             |                             |
| Field shift             | 0.6                         | 0.6                         | 1.2                         |
| Mass shift              | 0.4                         | 0.4                         | 0.9                         |
| Others                  |                             |                             |                             |
| Nuclear polarization    | 1.6                         | 1.1                         | 2.3                         |
| Nuclear deformation     | 1.5                         | -2.2                        | -2.4                        |
| Total IS theory         | -108.2(22)                  | -110.8(31)                  | -218.5(32)                  |

$1s^22s2p_{1/2}(J = 1) - 1s^22s^2$

| Main contributions      |                             |                             |                             |
| Field shift             | -112.8                      | -111.0                      | -221.3                      |
| Mass shift              | 0.1                         | 0.1                         | 0.2                         |
| QED                     |                             |                             |                             |
| Field shift             | 0.6                         | 0.6                         | 1.2                         |
| Mass shift              | 0.4                         | 0.4                         | 0.9                         |
| Others                  |                             |                             |                             |
| Nuclear polarization    | 1.6                         | 1.1                         | 2.3                         |
| Nuclear deformation     | 1.5                         | -2.2                        | -2.4                        |
| Total IS theory         | -108.5(22)                  | -111.1(31)                  | -219.1(32)                  |

*The uncertainty of $\delta \langle r^2 \rangle$ is not included.

To evaluate the nuclear deformation effect, one has to replace the standard spherically-symmetric Fermi model [3] for the nuclear charge distribution by [32]

$$\rho(r) = \frac{1}{4\pi} \int d\theta \rho(\theta),$$

(8)

where $\rho(r)$ is the axially-symmetric Fermi distribution

$$\rho(r) = \frac{N}{1 + \exp[(r - r_0(1 + \beta_{20}Y_{20}(\theta) + \beta_{40}Y_{40}(\theta))/a]}$$

(9)

consistent with the normalization condition $\int d\theta \rho(r) = 1$. Here $Y_{20}(\theta)$ and $Y_{40}(\theta)$ are the spherical functions, $\beta_{20}$ and $\beta_{40}$ are the quadrupole and hexadecapole deformation parameters [32–35]. The difference between the nuclear size effect obtained with the deformed model (8) and the standard spherically-symmetric Fermi model (3) at the same rms radius is ascribed to the nuclear deformation effect.

Finally, the interaction between the electrons and the nucleons causes the nucleus to make virtual transitions to excited states. This results in the increase of the binding energy of the electrons. To evaluate this effect, which is known as the nuclear polarization effect, one should consider the two-photon electron-nucleus interaction diagrams in which the intermediate nuclear states are excited. The calculations of this effect were performed in [36–39].

3. Results and discussion

In tables 1, 2 the individual contributions to the isotope shifts of the $1s^22s2p_{1/2}(J) - 1s^22s^2$ transition energies in $^{232,230}\text{Th}^{86+}$, $^{238,236}\text{U}^{88+}$ and $^{238,234}\text{U}^{88+}$ are presented. The nuclear charge radii and the $\delta \langle r^2 \rangle$ differences have been taken from [40]. The field shifts are calculated using the Dirac–Coulomb–Breit Hamiltonian using the formulas (5)–(6). The calculations are performed using the conformation-interaction Dirac–Fock–Sturm method for an extended nucleus [17]. The excited configurations are obtained from the basic configuration via single, double, and triple excitations of electrons. The accuracy of the calculations is defined by the stability of the results with respect to a variation of the basis size. In the present paper we use the basis including the excitations up to $(20s 20p 20d 15f 15g 15h 15i 15k 15l 15m 15n 15o)$. The contribution of quadrupole excitations was estimated in a smaller basis. It was found to be negligible for the ions under consideration. The same method has been used to calculate the
mass shifts within the approximation defined by the effective nuclear recoil operator (7).

The QED corrections to the field shifts have been evaluated in the one-electron approximation using the related formulas from [27, 28]. The QED effect on the mass shift was obtained as a sum of one- and two-electron contributions evaluated to zeroth order in 1/Ze. The one-electron terms have been taken from [31] while the two-electron corrections have been calculated in this work. As one can see from the tables, the QED recoil contribution is even larger than the mass shift obtained within the framework of the Breit approximation.

The nuclear deformation and polarization effects have been taken from the related calculations for Li-like thorium and uranium [87]. The uncertainties of these effects determine the total theoretical uncertainties. It should also be noted that the uncertainties of δ(r^2) are not included in the total uncertainties presented in tables 1 and 2. For checking purposes, instead of using equations (5)–(6), we have also performed the direct calculation of the field shift by taking the difference between the binding energies for the corresponding isotopes. We have found that in case of the 238.234U88+ isotope shift the direct calculation changes the result obtained using the F factor by 0.3 meV. This value, which includes also the effect of the higher-power-charge-radius change (apart from 〈r^2〉) [41], is small compared to the total theoretical uncertainty. However, the direct calculation of the field shift can be important when the isotope radius difference becomes rather large.

4. Conclusion

The isotope shifts of the 1s^2 2s 2p_j/2(J) → 1s^2 2s^2 transition energies in Be-like thorium and uranium ions are calculated including the relativistic, electron–electron correlation, Breit, and QED contributions. The nuclear polarization and nuclear deformation corrections are taken into account within the framework of the independent-electron approximation. The QED effects contribute on the level of the total uncertainty which is mainly defined by the nuclear polarization and deformation effects.

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