Hyperfine induced transition probabilities from $4f^{14}5s5p \, ^3P_{0,2}$ states in Sm-like ions

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Abstract

The hyperfine induced $4f^{14}5s5p \, ^3P_{0,2} - 4f^{14}5s^2 \, ^1S_0$ transition probabilities for highly charged Sm-like ions are calculated within the framework of the multiconfiguration Dirac–Hartree–Fock method. Electron correlation, the Breit interaction and quantum electrodynamical effects are taken into account. For ions ranging from $Z = 79$ to $Z = 94$, $4f^{14}5s5p \, ^3P_0$ is the first excited state, and the hyperfine induced transition (HIT) is a dominant decay channel. For the $4f^{14}5s5p \, ^3P_2$ state, the HIT rates of Sm-like ions with $Z = 82 - 94$ are reported as well as the magnetic dipole (M1) $^3P_2 - ^3P_1$, the electric quadrupole (E2) $^3P_2 - ^3P_0$, and the magnetic quadrupole (M2) $^3P_2 - ^1S_0$ transition probabilities. It is found that M1 transition from the $4f^{14}5s5p \, ^3P_2$ state is the most important decay channel in this range on $Z \geq 82$.

Keywords: hyperfine interaction, hyperfine induced transition, metastable states, multiconfiguration Dirac–Hartree–Fock method, electron correlation

(Some figures may appear in colour only in the online journal)

1. Introduction

The influence of the hyperfine interaction on lifetimes of metastable states was first noted by Bowen in a comment to a paper studying the ‘forbidden’ line of mercury by Huff and Houston [1]. The lifetimes of metastable states for the $nsnp \, ^3P_{0,2}$ states in the divalent atoms or ions were found to be significantly reduced by hyperfine induced transitions (HITs) [2, 3], especially for the $^3P_0$ state, because the single-photon $0 - 0$ transition is forbidden in the absence of hyperfine interactions. In the presence of a finite nuclear spin, the hyperfine interaction introduces the mixings of wave functions between the $nsnp \, ^3P_{0,2}$ and $1^3P_1$ levels, opening single-photon electric dipole transitions to the ground state $ns^2 \, ^1S_0$. This HIT attracts much attention in view of its potential applications, for instance, developing atomic clocks [4–6], diagnosing plasma parameters [7, 8] and determining nuclear parameters [9, 10]. Since the pioneering work of Garstang [11], there have been a number of theoretical studies of HITs of $nsnp \, ^3P_{0,2} - ns^2 \, ^1S_0$ in the Be-like ($n = 2$) [7, 12–15], the Mg-like ($n = 3$) [7, 16–18], and the Zn-like ($n = 4$) [19–21] isoelectronic sequences. On the experimental side, Brage et al used a planetary nebula to determine the HIT rates of $2s2p \, ^3P_0 - 2s^2 \, ^1S_0$ for Be-like $^{14}N$ and $^{15}N$ [8]. Schippers et al measured the lifetime of the $2s2p \, ^3P_0$ state for Be-like $^{47}Ti$ and $^{33}S$ [22, 23], Rosenband et al determined the lifetime of the $3s3p \, ^3P_0$ state for Mg-like $^{27}Al$ [6]. The detail about HIT can be found in the review by Johnson [2] and Grumer et al [3].

Here, we focus on the influence of HITs on lifetimes of the metastable $4f^{14}5s5p \, ^3P_{0,2}$ states in Sm-like ions. The ground state of neutral Sm ($Z = 62$) is $[Kr]4d^{10}5s^25p^6$ for Be-like and heavier ions [24]. The scheme of the low-lying levels calculated in the Dirac–Hartree–Fock (DHF) approximation for $W^{12+}$ is shown in figure 1. It is found that the spectra are much more complicated than other divalent atomic systems such as Mg- and Cd-like ion. There are 12 states for configuration $4f^{14}5s^25p$ and 171 states for $4f^{14}5s5p^2$, the energy levels of which
overlap with those in odd parity configuration \(4f^{14} 5s 5p\) due to the small energy difference between \(4f\), \(5s\), and \(5p\) orbitals. The \(4f^{14} 5s 5p^3 \Pi_{0,2}\) states are not as metastable as the \(5s 5p^3 \Pi_{0,2}\) states in other divalent atomic systems. However, the energy difference between \(4f\) and the outermost \(5s\) or \(5p\) orbitals increases with \(Z\), because the \(4f\) shell is more tightly bound for ions with the higher degree of ionization [24]. In figure 2, we present the calculated excitation energies of the \(4f^{14} 5s 5p^3 \Pi_{0,1,2}\), \(\Pi_1\) states along the Sm-like isoelectronic sequence, as well as the excitation energy of the \(4f^{13} 5s^2 5p^2 \Pi_3\) state, which is the lowest level with respect to configurations with open \(4f\) shell. The curve describing the excitation energies of the \(4f^{13} 5s^2 5p^2 \Pi_3\) state crosses that of the \(4f^{14} 5s 5p^3 \Pi_0\) state around \(Z = 79\), and also the one for the \(4f^{14} 5s 5p^3 \Pi_2\) state at about \(Z = 82\). Therefore, the \(4f^{14} 5s 5p^3 \Pi_0\) and \(\Pi_2\) levels become the metastable states when \(Z \geq 79\) and \(Z \geq 82\), respectively, and their lifetimes depend on probabilities of forbidden transitions.

In this work, we present an investigation on the hyperfine induced \(4f^{14} 5s 5p^3 \Pi_{0,2} - 4f^{14} 5s^2 5p^2 \Sigma_0\) transitions for Sm-like ions between \(Z = 79\) to \(Z = 94\). Meanwhile, we calculate rates of the magnetic dipole (M1) \(\Pi_2^0 - \Pi_1^0\), the electric quadrupole (E2) \(\Pi_2^0 - \Pi_0^1\), and the magnetic quadrupole (M2) \(\Pi_2^2 - \Sigma_0\) transitions, which are important decay channels for the \(4f^{14} 5s 5p \Pi_2\) level of ions with \(Z \geq 82\) besides the HIT.

2. Theory

2.1. The multiconfiguration Dirac–Hartree–Fock (MCDHF) method

Within the framework of the MCDHF method, atomic state functions (ASFs) are represented as a linear combination of symmetry-adapted configuration state functions (CFSs)

\[
|\GammaJM_\ell⟩ = \sum c_{\GammaJM_\ell} |\gammaJM_\ell⟩, \tag{1}
\]

where \(J\) and \(M\) are the total angular momentum and magnetic quantum numbers, respectively, \(\Gamma\) and \(\gamma\) are the additional quantum number defining each ASF or CSF uniquely. Configuration mixing coefficients \(c_{\GammaJM_\ell}\) are obtained through diagonalizing the Dirac–Coulomb Hamiltonian

\[
H_{DC} = \sum_{i=1}^{N} \left[ c_{\alpha_i} \cdot p_i + (\beta_i - 1) c^2 + V(r_i) \right] + \sum_{i<j}^{N} \frac{1}{r_{ij}}. \tag{2}
\]

Here, \(V(r_i)\) is the monopole part of the electron-nucleus Coulomb interaction, and \(\alpha_i\) and \(\beta_i\) are the Dirac matrices. In the relativistic self-consistent field procedure, both the radial parts of Dirac orbitals and the mixing coefficients \(c_{\GammaJM_\ell}\) are optimized for minimizing the energy of the atomic states concerned [25]. The Breit interaction in the low-frequency approximation and quantum electrodynamical (QED) effects including the vacuum polarization and the self-energy correction can be included in the subsequent relativistic configuration interaction (RCI) calculation [26–28].

2.2. HITs

Hyperfine interactions couples nuclear spin \(I\) and electronic angular momentum \(J\) to total angular momentum \(F\), and only the \(F\) and \(M_F\) are good quantum numbers. The wave function of a hyperfine level can be written as

\[
|FM_F⟩ = \sum_{I,J} h_{IJ,F} |I,JFM_F⟩, \tag{3}
\]

where \(h_{IJ,F}\) are the hyperfine mixing coefficients, and can be obtained in first-order perturbation theory by

\[
h_{IJ,F} = \frac{|I,JFM_F⟩ ⟨H_{\text{interaction}}⟩ _{IJ,F}⟩ F_{IJ,F} - E_{IJ,F}}{E_{IJ,F} - E_{IJ,F}^0}. \tag{4}
\]

Here, the subscript 0 labels the unperturbative level. The
The hyperfine interaction Hamiltonian $H_{hf}$ is expressed as
\[ H_{hf} = \sum_k T^{(k)} \cdot M^{(k)}, \]
where $T^{(k)}$ and $M^{(k)}$ are spherical tensor operators of rank $k$ acting on the electronic and nuclear parts of the wave function, respectively. The hyperfine interaction matrix elements can be expressed in terms of reduced electronic and nuclear matrix elements as [20]
\[ \langle \Gamma J' \rangle [T^{(k)} \cdot M^{(k)}] = \langle \gamma_0 J_0 \Gamma J' | T^{(k)} | \gamma_0 J_0 \Gamma J' \rangle. \]

In this work, only the magnetic dipole ($k = 1$) and the electric quadrupole ($k = 2$) hyperfine interactions are included. The reduced matrix elements of the nuclear tensor operators are related to nuclear magnetic dipole moment $\mu_I$ and electric quadrupole moment $Q$ through
\[ \langle I|M^{(1)}||I\rangle = \mu_I \sqrt{1 + F^{-1},} \]
and
\[ \langle I|M^{(2)}||I\rangle = Q \frac{2}{(2I + 3)(I + 1)} \sqrt{\frac{I(I + 1)}{2(I - 1)}}. \]

The probability $A$ (in s$^{-1}$) for an electric dipole (E1) transition between two hyperfine levels can be written as [14]
\[ A = \frac{2.02613 \times 10^{18}}{\lambda^2(2F + 1)} [\langle \Gamma J I | D^{(1)} || \Gamma' J' F' \rangle]^2. \]

Here, $\lambda$ is wavelength in Å, and $D^{(1)}$ is the electric dipole tensor operator. The reduced matrix elements can be further simplified to
\[ \langle \Gamma J F || D^{(1)} || \Gamma' J' F' \rangle = \sqrt{2(F + 1)(2F + 1)} \times (-1)^{F + J + F' + 1} \left\{ \begin{array}{ccc} J & F & I \\ F' & J' & 1 \end{array} \right\} \langle \Gamma J || D^{(1)} || \Gamma' J' \rangle. \]

Using equations (4), (10) and (11) the transition probability is given by
\[ A = \frac{2.02613 \times 10^{18}}{\lambda^2}(2F + 1) \sum_{J \in \Omega} \sum_{J' \in \Omega} \sum_{\Omega} h_{\Omega J} h_{\Omega' J'} \left\{ \begin{array}{ccc} J & F & I \\ F' & J' & 1 \end{array} \right\} [\langle \Gamma J || D^{(1)} || \Gamma' J' \rangle]^2. \]

In practical calculation, we only consider the hyperfine interactions between the states in the same configuration. For example, the hyperfine level belonging to the $4f^{14} 5s^2 3p^0_0$ state is given by
\[ \langle \Gamma J F || M^{(1)} || \Gamma' J' F' \rangle^{(1)} = \langle \Gamma J F || M^{(1)} || \Gamma' J' F' \rangle^{(0)} + \sum_{S=0,1} h_{\Sigma J} h_{\Sigma' J} \langle \Gamma J || D^{(1)} || \Gamma' J' \rangle. \]

The mixing between the $4f^{14} 5s^5 3p^{0,2}_0$ and $^{(2S+1)}P^{0,2}_S$ ($S = 0, 1$) states gives rise to a single-photon E1 transition from the $4f^{14} 5s^5 3p^{0,2}_0$ levels to the $4f^{14} 5s^2 1S_0$ ground state. The corresponding hyperfine induced $4f^{14} 5s^5 3p^{0,2}_0 - 4f^{14} 5s^2 1S_0$ transition rates can be given by
\[ A_{HF}(5s5p \quad ^pP^0_0 - 5s^2 1S_0) = \frac{2.02613 \times 10^{18}}{33} \left| \sum_S h_{\Sigma J} h_{\Sigma' J} \langle \Gamma J || D^{(1)} || \Gamma' J' \rangle \right|^2. \]

Since the hyperfine mixing coefficients depend on nuclear parameters, it is difficult to investigate the trend of the HIT rates along the isoelectronic sequence. Therefore, we define a reduced HIT rate [7, 14, 18]
\[ A_{el} = \frac{A_{HF}}{\mu_f^2(1 + I^{-1})(2I + 1)} \times \left( W(J_{el}J_{el}, F_{el}) \right)^2. \]

Here, $W(J_{el}J_{el}; F_{el})$ is the 6j-symbol as in equation (7) and $A_{HF}$ indicates that only the magnetic dipole hyperfine interaction was taken into account in $h_{\Sigma J} h_{\Sigma' J}$. It is clear that $A_{el}$ is independent of the nuclear parameters.

### 2.3. Computational model

The levels belonging to configuration $4f^{14} 5s^2$ and $4f^{14} 5s^5p$ are optimized in two separate MCDHF calculations. The configuration space is expanded by employing the active space approach. We start from the DHF calculations, in which the occupied orbitals are optimized as spectroscopic. In order to take into account the correlations between valence orbitals $4f$, $5s$ and $5p$, CSFs generated by single and double (SD) excitations from the valence orbitals to the virtual orbitals are included into the configuration space. Due to stability problems in the relativistic self-consistent field procedure, the virtual orbitals are augmented layer by layer up to $n = 8$, and only the outermost layer is optimized in each step. Each virtual orbital layer contains orbitals with the $s$, $p$, $d$, $f$ and $g$ symmetries. In table 1, the virtual orbitals are listed by angular symmetry and the number of orbitals for each symmetry are enclosed in quotation marks to avoid confusion with spectroscopic orbitals. For example, '2spdfg' stands for two $s$, $p$, $d$, $f$ and $g$ virtual orbitals. The core-valence (CV) and core-core (CC) correlations involving $4s$, $4p$ and $4d$. 

| Model | $^3P^0_0$ | $^3P^0_1$ | $^3P^2_0$ | $^3P^2_1$ |
|-------|---------|---------|---------|---------|
| DHF   | 285869  | 31110   | 476907  | 551998  |
| '1spdfg' | 296863  | 31945   | 488005  | 549689  |
| '2spdfg' | 299017  | 320578  | 490166  | 549180  |
| '3spdfg' | 299292  | 320731  | 490367  | 548763  |
| + CV & CC | 297896  | 318459  | 489498  | 548367  |
| + Bret | 299450  | 320918  | 489215  | 548204  |
| + QED | 296660  | 318132  | 486483  | 545359  |
| Curtis | 285669  | 310705  | 474659  | 549501  |
| Exp. [34] | 318878  |         |         |         |

* Is converted from the measured wavelength 31.36 nm.

| Table 1. Calculated excitation energies (in cm$^{-1}$) of $4f^{14} 5s5p \quad ^pP^S_0$ states for the Sm-like Au ion. DHF represents the uncorrelated DHF calculation. 'nspdfg' stands for the virtual orbital set. CV & CC indicate RCI computations taking CV and CC correlations into account. |
shells are taken into account in the following RCI calculations. In this step, CFs generated by SD excitations from these three core orbitals to all virtual orbitals are included. This step resulted in the calculations converging, but in order to include spin-polarization in deep s-subshell [31, 32] we add the configurations generated by single excitations from 1s, 2s and 3s, to virtual s-orbitals in the last step. Furthermore, the Breit interaction and QED effects are considered in the RCI computations.

3. Results and discussion

Taking Au$^{17+}$ ($Z = 79$) as an example, we present in table 1 the excitation energies of the 4$f$$^{14}$ 5s5p 1$^3P_1$ states as functions of the computational models, as well as the available experimental and theoretical values. In Curtis’ calculation [33], only the configurations 4$f$$^{14}$ 5s$^2$, 4$f$$^{14}$ 5s5p, 4$f$$^{14}$ 5s$^2$5p, and 4$f$$^{14}$ 5s5p$^2$ are included, and the excitation energy of 4$f$$^{14}$ 5s5p 3$^3P_1$ is about 8000 cm$^{-1}$ lower than the experimental value [34]. The present multiconfiguration calculation gives much better results. It was found that the main valence correlations were captured by three virtual orbital layers, which make a larger contribution to the excitation energies than the CV and CC correlations. The effects of the Breit interaction and QED on the excitation energies are similar to the CV and CC correlations.

Table 2 presents the line strengths of the 1$^1P_1 \rightarrow 1S_0$ E1 transitions in the Babushkin and Coulomb gauges for Au$^{17+}$, respectively. Convergence trends of the line strengths are similar to the excitation energies. In addition, with the expansion of the configuration space, the agreement between the two gauges is significantly improved. After including the Breit interaction and QED effects, the difference in line strengths between these two gauges is only about 2% for the 1$^1P_1 \rightarrow 1S_0$ transition and 8% for the 3$^3P_1 \rightarrow 1S_0$ transition. The line strength in the Babushkin gauge is used to calculate the HIT rates, since it is relatively insensitive to electron correlation effects [35].

3.1. Hyperfine induced 4$f$$^{14}$ 5s5p 3$^3P_0 - 4f$$^{14}$ 5s$^2$ 1$S_0$ transition probability

In table 3, the hyperfine induced 4$f$$^{14}$ 5s5p 3$^3P_0 - 4f$$^{14}$ 5s$^2$ 1$S_0$ transition probabilities (A_{HIT}) and corresponding transition probabilities (A_{HIT}) for Sm-like ions with $Z = 79 - 94$. The uncertainty in the nuclear parameters is given in parentheses.

![Figure 3. Rates in s$^{-1}$ of hyperfine induced 3$^3P_0 \rightarrow 1S_0$ electric dipole transition $A_{HIT}$ and reduced HIT $A_{HIT}$ electric dipole transition $A_{HIT}$ and reduced HIT $A_{HIT}$ as well as the fitting curve by using equation (14).](image)

![Table 2. Line strengths of the 1$^1P_1 \rightarrow 1S_0$ E1 transitions for the Sm-like Au ion. S_C and S_B stand for the line strengths in Babushkin and Coulomb gauges, respectively.](table2)

| Model      | S_C | S_B | S_C | S_B |
|------------|-----|-----|-----|-----|
| DHF        | 1.028 | 1.274 | 0.165 | 0.237 |
| 1$^3pdgs'$ | 0.959 | 1.000 | 0.194 | 0.202 |
| 2$^3pdgs'$ | 0.909 | 0.944 | 0.189 | 0.198 |
| 3$^3pdgs'$ | 0.895 | 0.930 | 0.186 | 0.196 |
| + CV & CC  | 0.902 | 0.933 | 0.179 | 0.199 |
| +Breit     | 0.907 | 0.937 | 0.180 | 0.198 |
| +QED       | 0.919 | 0.937 | 0.184 | 0.198 |

![Table 3. Transition energies ($\Delta E$) in cm$^{-1}$, probabilities ($A_{HIT}$) and reduced rates ($A_{HIT}$) in s$^{-1}$ of HIT 4$^{14}$ 5s5p 3$^3P_0 - 4f$$^{14}$ 5s$^2$ 1$S_0$ for Sm-like ions with $Z = 79 - 94$.](table3)
between the $^3P_0$ and $^1P_1$ states. Hence, the HIT rate can be expressed as

$$A_{\text{HT}}(^3P_0 - ^1S_0) = \mu_1^2 (1 + I^{-1}) A_{cl}(^3P_0 - ^1S_0), \quad (15)$$

which factorizes the HIT rate into the nuclear and electronic parts. The reduced HIT rates $A_{cl}$ are independent of the nuclear parameters and have a smooth behavior along the isoelectronic sequence. Furthermore, the reduced HIT rates can be fitted as a power function of $Z$ as

$$A_{cl}(^3P_0 - ^1S_0) = 1.0198 \times 10^{-27} Z^{1.4569}. \quad (16)$$

The fitting curve is also depicted in figure 3, which is in good agreement with the \textit{ab initio} calculation.

### 3.2. Decay of the $4f^{14}5s5p^2P_2$ state

The HIT probabilities ($A_{\text{HT}}$) of the $^3P_0^u$ level and corresponding transition energies ($\Delta E$) for Sm-like ions with $Z = 82 - 94$ are presented in Table 4. Apart from the HIT, the $^3P_2$ state can decay through the magnetic dipole (M1) transition ($^3P_2 - ^1P_0^u$), the electric quadrupole (E2) transitions ($^3P_2 - ^1P_0^u$), and the magnetic quadrupole (M2) transition ($^3P_2 - ^1S_0$). To show the competition among these decay channels, the trend of the reduced HIT probabilities as well as the M1, M2 and E2 transition probabilities of this state along the isoelectronic sequence are illustratively presented in figure 4. The HIT probabilities for the isotopes of each element concerned are plotted with scattering symbols. It is found that the M1 transition is dominant for these ions, and the HIT probabilities have the same order of magnitude as those two E2 transitions ($^3P_2 - ^1P_0^u$), but also depend on the nuclear parameters.

As mentioned above, the electric quadrupole hyperfine interaction is not taken into account in $A_{cl}$. In order to show the contribution from the electric quadrupole hyperfine interaction to the HIT rate for the $^3P_2^u$ state, the ratios between the HIT probabilities $A_{\text{HT}}$ and the rates $A_{\text{HT}}^{M1}$, where the latter only includes the magnetic dipole hyperfine interaction, are presented in figure 5. It is clear that the electric quadrupole hyperfine interaction is important to the HIT rate only for isotopes with larger nuclear electric quadrupole moment $Q$ compared with magnetic dipole moment $\mu_1$. For example, for $^{229}$Th with $Q = 4.3(9)$ and $\mu_1 = 0.46(4)$ the electric

| Nucleus | $I$ | $\mu_1$ | $Q$ | $\Delta E$ | $A_{cl}$ | $F$ | $A_{\text{HT}}$ |
|---------|-----|---------|-----|------------|----------|-----|----------------|
| $^{205}$Pb | 5/2 | 0.7117(4) | 0.23(4) | 592380 | 6.188[3] | 1/2 | 0 |
| $^{209}$Po | 1/2 | 0.68(8) | | 671560 | 1.065[4] | 3/2 | 8.209[2] |
| $^{209}$Rn | 5/2 | 0.8388(4) | 0.31(3) | 756856 | 1.804[4] | 1/2 | 0 |
| $^{211}$Ra | 5/2 | 0.878(4) | 0.46(5) | 852112 | 3.045[4] | 1/2 | 0 |
| $^{229}$Th | 5/2 | 0.46(4) | 4.3(9) | 956641 | 5.105[4] | 1/2 | 0 |
| $^{233}$U | 5/2 | 0.59(5) | 3.663(8) | 1072110 | 8.527[4] | 1/2 | 0 |
| $^{241}$Pu | 5/2 | 0.683(15) | 6(2) | 1197904 | 1.416[5] | 1/2 | 0 |
quadrupole hyperfine interaction changes the HIT rate \( (F = 3/2) \) by a factor of about 2. For isotope \(^{209}\)Po with nuclear spin \( I = 1/2 \), however, the HIT only depends on the magnetic dipole hyperfine interaction.

4. Summary

With increasing atomic number \( Z \), the ground state of Sm-like ions become \([\text{Kr}]4d^{10}4f^{14}5s^2 \, ^1S_0\), and the \( 4f^{14}5s5p \, ^3P_0 \) and \(^3P_2\) states turn into metastable states for the ions with \( Z \gtrsim 79 \) and \( Z \gtrsim 82 \), respectively. Using the multiconfiguration DHF method, we calculate the hyperfine induced \( 4f^{14}5s5p \, ^3P_0 - 4f^{14}5s^2 \, ^1S_0 \) transition rates for these ions. For the \( 4f^{14}5s5p \, ^3P_2 \) state, the probabilities of other important decay channels including the M1, M2 and E2 transitions are also reported. For the first excited state \( 4f^{14}5s5p \, ^1P_1 \), the HIT is a dominant single-photon decay channel, and thus significantly impacts the lifetime of this state. A fitting formula in \( Z \) for the electronic part of the HIT rate is further derived in order to estimate the HIT rate for any isotope along the isoelectronic sequence. For the other metastable state, it is shown that the M1 transition is the most important decay channel for these ions with \( Z \gtrsim 82 \).

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