

CALCULATION OF THALLIUM HYPERFINE ANOMALY*

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Abstract. We suggest a method of a computation of hyperfine anomaly for many-electron atoms and ions. At first, we tested this method by calculating the hyperfine anomaly for a hydrogen-like thallium ion and obtained fairly good agreement with analytical expressions. Then, we did calculations for the neutral thallium and tested an assumption that the ratio between the anomalies for *s* and *p*_{1/2} states is the same for these two systems. Finally, we come up with recommendations about the preferable atomic states for the precise measurements of the nuclear *g* factors.

Key words: Hyperfine anomaly, hyperfine structure, thallium, nuclear distribution, Bohr–Weisskopf effect

1. INTRODUCTION

In recent years, the precision achieved in resonant ionization spectroscopy experiments coupled with advances in atomic theory has enabled new atomic physics based tests of nuclear models. Understanding the occurrence of shape coexistence in atomic nuclei is one of them. This phenomenon is associated with the existence of both the near-spherical and deformed structures of nuclei for neutron-deficient isotopes near the value of *Z* = 82 closed shell [1]. The measurements of hyperfine constants and isotope shifts are highly sensitive to the changes of the nuclear charge and magnetic radii because they depend on the behavior of the electron wave function near the nucleus. The hyperfine structure (HFS) splitting measurements can serve as a very useful tool for the understanding of the shape coexistence phenomena in atomic nuclei.

Magnetic hyperfine constants *A* are usually assumed to be proportional to the nuclear magnetic moments. However, this is true only for the point-like nucleus. For the finite nucleus, we need to take into account (i) distribution of the magnetization inside the nucleus and (ii) dependence of the electron wave function on the nuclear charge radius. Former correction is called magnetic (Bohr–Weisskopf) [2] and the latter is called charge correction (Breit-Rosenthal) [3, 4]. Together, these corrections are known as the hyperfine anomaly [5]. Below, we discuss how to calculate the hyperfine anomaly for many-electron atoms with the available atomic packages. We use a thallium atom as a reference system for our calculations, because there are comprehensive experimental data [6-10] and many theoretical calculations for this atom [5, 11-14].

Shabaev [5] and Shabaev et al. [11] found the analytical expressions for the hyperfine anomaly for H-like thallium ion. For the neutral thallium, there is the numerical calculation by Mårtensson-Pendrill [12].

Experimentally, HFS anomaly is studied much better for a neutral Tl than for a respective H-like ion. In the work [15], it has been suggested that the ratio between the anomalies for *s* and *p*_{1/2} states remains constant for these two systems. Here we try to test this assumption.

We use the atomic package [16], which is based on the original Dirac-Hartree-Fock code [17]. This package is often used to calculate different atomic properties including hyperfine structure constants of Tl [13,14], Yb [18], Mg [19], and Pb [20].

2. THEORY AND METHODS

A four component Dirac wave function of an electron in a spherically symmetric atomic potential can be written as [17]:

$$\psi_{n,\kappa,m}(\mathbf{r}) = \frac{1}{r} \begin{pmatrix} P_{n,\kappa}(r)\Omega_{\kappa,m}(\omega) \\ -Q_{n,\kappa}(r)\Omega_{-\kappa,m}(\omega) \end{pmatrix}, \quad (1)$$

where the relativistic quantum number $\kappa=(l-j)(2j+1)$ and $\Omega_{\kappa,m}$ is the spherical spinor. In these notations, the radial integral for the magnetic hyperfine constant for the point-like nuclear magnetic moment in the origin has the form:

$$I_{n',\kappa',n,\kappa} = \int_0^\infty (P_{n',\kappa'} Q_{n,\kappa} + Q_{n',\kappa'} P_{n,\kappa}) \frac{dr}{r^2}. \quad (2)$$

In the case of uniformly distributed magnetic moment over the nucleus of radius *R*_N the part of this radial integral inside the nucleus modifies to [12]:

$$I_{n',\kappa',n,\kappa}^{muc} = \int_0^{R_N} (P_{n',\kappa'} Q_{n,\kappa} + Q_{n',\kappa'} P_{n,\kappa}) \frac{rdr}{R_N^3}. \quad (3)$$

Outside the nucleus we can still use the integrand from Eq. (2).

In our package, we use the model of the uniformly charged ball and, inside the nucleus we use Taylor expansion for the radial functions *P* and *Q*:

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$$P_{n,\kappa}(r)|_{r \leq R_N} = r^{|\kappa|} \sum_{k=0}^M P_{n,\kappa,k} x^k, \quad x = \frac{r}{R_N}. \quad (4)$$

With the help of this expansion, we can calculate the integral (3):

$$I_{n',\kappa',n,\kappa}^{nuc} = R_N^{|\kappa'|+|\kappa|-1} \sum_{m=0}^M \sum_{k=0}^m \frac{P_{n',\kappa',k} Q_{n,\kappa,m-k} + Q_{n',\kappa',k} P_{n,\kappa,m-k}}{|\kappa'|+|\kappa|+m+2}. \quad (5)$$

Note that nuclear contribution to the integral (2) can be written as:

$$I_{n',\kappa',n,\kappa}^{nuc,0} = R_N^{|\kappa'|+|\kappa|-1} \sum_{m=0}^M \sum_{k=0}^m \frac{P_{n',\kappa',k} Q_{n,\kappa,m-k} + Q_{n',\kappa',k} P_{n,\kappa,m-k}}{|\kappa'|+|\kappa|+m-1}. \quad (6)$$

Using expression (6) for two different nuclear radii, we can calculate the charge correction to atomic HFS, while using the expression (5) we simultaneously account for the charge and magnetic corrections. However, these two models are not sufficiently flexible to accurately describe the hyperfine anomaly. Expressions (2) and (3) correspond to two variants of the distribution of the magnetic moment: either homogeneous distribution over the whole nucleus, or point-like dipole in the origin. Introducing the magnetic radius R_M , we get either $R_M = R_N$ or $R_M = 0$. If we want to allow for the arbitrary value $R_M \leq R_N$, we need to combine the integrand from Eq. (3) for $r < R_M$ with the integrand from Eq. (2) for $R_M \leq r \leq R_N$. Then we can write the integral over the nucleus as

$$I_{n',\kappa',n,\kappa}^{nuc}(R_N, R_M) = I_{n',\kappa',n,\kappa}^{nuc}(R_M) + \left(I_{n',\kappa',n,\kappa}^{nuc,0}(R_N) - I_{n',\kappa',n,\kappa}^{nuc,0}(R_M) \right), \quad (7)$$

$$I_{n',\kappa',n,\kappa}^{nuc}(R_M) = R_M^{|\kappa'|+|\kappa|-1} \sum_{m=0}^M \sum_{k=0}^m \frac{P_{n',\kappa',k} Q_{n,\kappa,m-k} + Q_{n',\kappa',k} P_{n,\kappa,m-k}}{|\kappa'|+|\kappa|+m+2} \left(\frac{R_M}{R_N} \right)^m, \quad (8)$$

$$I_{n',\kappa',n,\kappa}^{nuc,0}(R_M) = R_M^{|\kappa'|+|\kappa|-1} \sum_{m=0}^M \sum_{k=0}^m \frac{P_{n',\kappa',k} Q_{n,\kappa,m-k} + Q_{n',\kappa',k} P_{n,\kappa,m-k}}{|\kappa'|+|\kappa|+m-1} \left(\frac{R_M}{R_N} \right)^m. \quad (9)$$

2.1. Isotope effect for magnetic HFS

Suppose we want to compare hyperfine constants A_1 and A_2 for two isotopes with nuclear g factors $g^{(1)}_I$ and $g^{(2)}_I$ ($g_I = \mu/I$), nuclear charge radii $R_N^{(1)}$ and $R_N^{(2)}$, and magnetic radii $R_M^{(1)}$ and $R_M^{(2)}$. We can write:

$$\frac{A_1}{A_2} = \frac{g^{(1)}_I}{g^{(2)}_I} \left(1 - \lambda^C \frac{R_N^{(1)} - R_N^{(2)}}{R_N^{(1)} + R_N^{(2)}} - \lambda^M \frac{R_M^{(1)} - R_M^{(2)}}{R_M^{(1)} + R_M^{(2)}} \right). \quad (10)$$

The anomaly then has the following form:

$$1 \Delta^2 \equiv \frac{g^{(2)}_I A_1}{g^{(1)}_I A_2} - 1 = - \left(\lambda^C \frac{R_N^{(1)} - R_N^{(2)}}{R_N^{(1)} + R_N^{(2)}} + \lambda^M \frac{R_M^{(1)} - R_M^{(2)}}{R_M^{(1)} + R_M^{(2)}} \right) \quad (11)$$

By solving above equations for several radii, we can find λ^C and λ^M and calculate the anomaly for the isotopes of interest. Below we will see that parameters λ^C and λ^M themselves depend on the radii R_N and R_M . Therefore, it is better to use parameters b_N and b_M defined below (see Eq. (18)).

2.2. Hydrogen-like ions

It is generally accepted that the observed hyperfine constant $A(R_N, R_M)$ of a one-electron ion may be presented in the following form:

$$A(R_N, R_M) = A_0 (1 - \delta(R_N)) (1 - \epsilon(R_M)). \quad (12)$$

Here $A_0 \equiv A(0,0)$ is the factor which is independent of the nuclear radii and $\delta(R_N)$ and $\epsilon(R_M)$ are the nuclear charge distribution and magnetic distribution corrections respectively. For a given Z and electron state, they can be written as:

$$\delta(R_N) = b_N R_N^{(2\gamma-1)}, \quad \epsilon(R_M) = b_M R_M^{(2\gamma-1)}, \quad (13)$$

where b_N and b_M are the factors which are independent of the nuclear radii, $\gamma = \sqrt{\kappa^2 + (\alpha Z)^2}$, and α is the fine structure constant. The expression for A_0 was obtained in the analytical form as [5]:

$$A_0 = \frac{\alpha(\alpha Z)^3 g_I}{j(j+1)} \frac{m}{m_p} \frac{\kappa(2\kappa(\gamma+n_r) - N)}{N^4 \gamma(4\gamma^2 - 1)} mc^2. \quad (14)$$

Here, m and m_p are the electron and proton masses, g_I is the nuclear g factor, j is the total electron angular momentum, $N = \sqrt{n_r^2 + 2n_r^2 \gamma + \kappa^2}$, n_r is the radial quantum number.

It follows from Eqs. (12) and (13) that if we calculate HFS constant numerically for different R_N and R_M , we should get the following dependence on the radii:

$$A(R_N, R_M) = A_0 (1 - b_N R_N^{2\gamma-1}) (1 - b_M R_M^{2\gamma-1}) \quad (15)$$

This expression defines the dependence of parameters λ^C and λ^M from (10) on the radii R_N and R_M . For example, on the one hand, we have:

$$\frac{A(R_N + \rho, R_M)}{A(R_N - \rho, R_M)} = 1 - \lambda^C(R_N) \frac{\rho}{R_N}. \quad (16)$$

On the other hand:

$$\frac{A(R_N + \rho, R_M)}{A(R_N - \rho, R_M)} = 1 + 2\rho \frac{\partial A(R_N, R_M) / \partial R_N}{A(R_N, R_M)}. \quad (17)$$

Then, from Eq. (16) we get:

$$\lambda^C(R_N) \approx \frac{2(2\gamma-1)b_N R_N^{2\gamma-1}}{1 - b_N R_N^{2\gamma-1}} \approx 2(2\gamma-1)b_N R_N^{2\gamma-1}. \quad (18)$$

Similar expressions can be obtained for $\lambda^M(R_M)$. For the point-like magnetic dipole approximation ($R_M = 0$) the magnetic correction is equal to zero, and the hyperfine constant can be fitted by the function:

$$A(R_N, 0) = A_0 (1 - b_N R_N^{2\gamma-1}). \quad (19)$$

For the uniform distribution of the charge and magnetic moment with the value of $R_M = R_N$ we get:

$$A(R_N, R_N) = A_0 (1 - (b_N + b_M) R_N^{2\gamma-1}). \quad (20)$$

2.3. Many-electron atoms

Since the one-electron radial integrals are defined, we can calculate the atomic HFS using many-electron wave functions and account for electronic correlations as described in Ref. [13]. Using Eqs. (7–9), we can calculate the atomic HFS constants for arbitrary radii R_N and R_M with the only constraint that $R_N \geq R_M$. We

can do configuration interaction calculations with the frozen core and few valence electrons. Then we can add the core-valence correlation corrections with the help of the many-body perturbation theory. At this stage, we substitute the valence radial integrals with the effective ones, which account for the spin polarization of the core. The latter are obtained by solving random-phase approximation (RPA) equations.

Effective radial integrals may have significantly different dependence on the parameters of the nucleus than initial “bare” integrals. This is particularly true for the orbitals with the high angular momentum. Because of the centrifugal barrier, these orbitals do not penetrate inside the nucleus and bare radial integrals do not depend on the nuclear size. On the other hand, spin-polarization of the core always includes polarization of the core s and $p_{1/2}$ shells. Because of that, all effective radial integrals are sensitive to the nuclear charge and magnetic distributions. In general, we can divide all correlation corrections in two classes: corrections, which mix orbitals within one partial wave, and the ones which mix different partial waves. For example, the self-energy type corrections belong to the first class. They mix core and valence orbitals of the same symmetry and can significantly change the orbital density at the origin. Therefore, these corrections change the size of the HFS matrix elements. On the other hand, all orbitals of the same symmetry have practically the same sensitivity to the nuclear distributions. Thus, such correlation corrections do not affect parameters b_N and b_M and the HFS anomaly (11). RPA corrections belong to the second class, which significantly contribute to the HFS anomaly.

Measurement of hyperfine anomaly for highly charged ions allows experimental study of QED effects (see e.g. [21]). Attempts to take into account QED corrections in hyperfine structure calculations for such systems have already been made [8, 22, 23]. However, for neutral atoms QED corrections are sufficiently small and should be taken into account only after correlation effects. It can be done using the method recently described in the article [24]. Note, that dominant QED corrections belong to the first class and do not significantly affect the HFS anomaly.

3. RESULTS AND DISCUSSION

3.1. HFS anomaly for H-like thallium ion

In this section, we calculate the HFS constants of the $1s$, $2s$, and $2p_{1/2}$ states of Tl^{80+} for different radii R_N and R_M and compare our results with the analytical expressions from Ref. [5]. Figure 1 shows the dependence of the hyperfine constant $A(1s)$ on the radii R_N and R_M . We see very good agreement with Eqs. (19) and (20).

Table I summarizes our results for H-like Tl ion. We see the perfect agreement of the calculated and analytical values of A_0 for all three states. Charge and magnetic corrections δ and ε were calculated in Ref. [11] for the $1s$ state of the isotope ^{203}Tl . These analytical values are also in a good agreement with our numerical results.

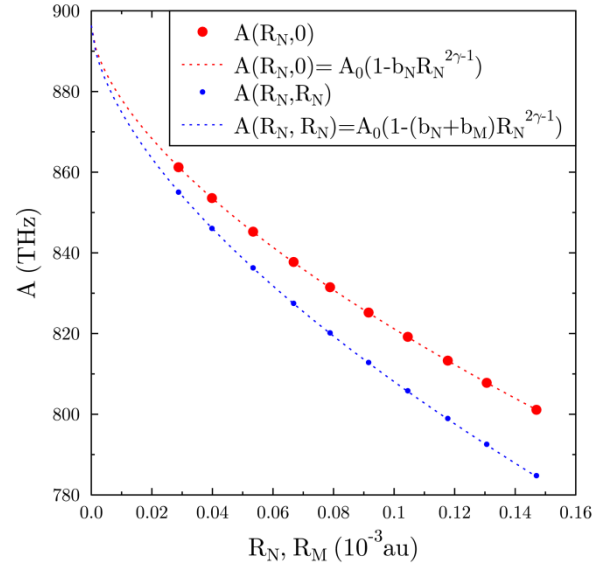


Figure 1. The dependence of the HFS constant $A(R_N, R_M)$ for the ground state of H-like Tl ion from nuclear charge and magnetic radii. Dots and circles correspond to the computed values. Dashed lines correspond to the fits by Eqs. (19) and (20).

Table 1. Compilation of the fitting parameters for HFS of H-like Tl ion: A_0 is HFS constant for point-like nucleus, δ and ε are the nuclear charge and magnetization distribution corrections parametrized by b_N and b_M coefficients respectively. We use g factor $g_I = 3.27640$. Corrections δ and ε for ^{203}Tl are calculated for $R_N = R_M = 0.1306 \times 10^{-3}$ au.

		$1s$	$2s$	$2p_{1/2}$
A_0 (THz)	fit.	896.4	144.9	45.0
	Eq. (14)	895.7	144.8	45.0
b_N	fit.	0.3441	0.3671	0.0960
	Ref. [11]	0.0988	–	–
$\delta(^{203}Tl^{80+})$	fit.	0.0988	0.1054	0.0276
	Ref. [11]	0.0988	–	–
b_M	fit.	0.0599	0.0638	0.0176
	Ref. [11]	0.0172	0.0183	0.0051
$\varepsilon(^{203}Tl^{80+})$	fit.	0.0172	0.0183	0.0051
	Ref. [11]	0.0179	–	–

Figure 2 shows how parameters λ for the $1s$ state depend on the radii R_N and R_M . We see the perfect agreement with the analytical expression (18). On the other hand, it means that these parameters strongly depend on the nuclear size. Because of that, they cannot be treated as a constant even for the isotopes with the similar radii. Therefore, it is better to use parameters b_N and b_M defined by Eq. (15).

According to our calculations (see Table I), the ratios of the parameters b_N and b_M for $1s$ and $2s$ states are close to unity: $\frac{b_N(1s)}{b_N(2s)} = 0.937$ and $\frac{b_M(1s)}{b_M(2s)} = 0.939$. This is expected, as wave functions of the same symmetry should be proportional to each other inside the nucleus. Similar ratios for $1s$ and $2p_{1/2}$ states are $\frac{b_N(1s)}{b_N(2p_{1/2})} = 3.58$ and $\frac{b_M(1s)}{b_M(2p_{1/2})} = 3.40$. Again, one can expect that these ratios only weakly depend on the principle quantum numbers.

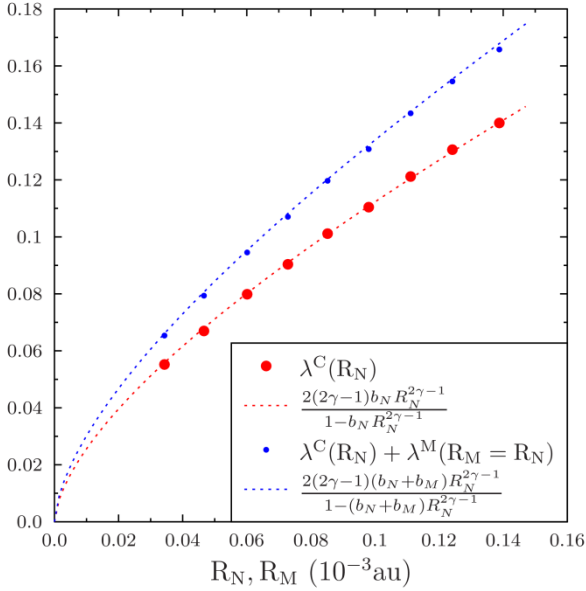


Figure 2. Dependence of the parameters $\lambda^C(R_N)$ and $\lambda^M(R_M)$ (see Eq. (10)) on the charge and magnetic radii of the nucleus for the ground state of H-like Tl ion. Computed values represented by points. The curves correspond to the fits with Eq. (18).

3.2. HFS anomaly of neutral thallium atom

The ground configuration of the neutral thallium is $[1s^2 \dots 6s^2]6p$ and the ground multiplet includes two levels, $6p_{1/2}$ and $6p_{3/2}$. The lowest level of the opposite parity is $7s$. Most of the experiments and calculations of the HFS in neutral thallium deal with these three levels. If we treat thallium as a one-electron system with the frozen core $[1s^2 \dots 6s^2]$, we can do the calculation using the Dirac-Hartree-Fock (DHF) method. In this case, the dependence of the HFS constants on the nuclear radii is similar to the one-electron ion.

In DHF approximation, the HFS constant with the value of $A(6p_{3/2}) = 1.30$ GHz is very small and practically does not depend on R_N and R_M (see Table II). At the same time, the HFS constants $A(6p_{1/2})$ and $A(7s)$ are well described by Eqs. (19, 20) (see Fig. 3). According to our calculations, the ratios between the coefficients b_N and b_M for s and $p_{1/2}$ waves are close to the respective ratios in the H-like ion. For example, the ratios of these constants for $1s$ state of the ion and $7s$ state of the neutral atom are $\frac{b_N(1s)}{b_N(7s)} = 0.926$ and $\frac{b_M(1s)}{b_M(7s)} = 0.931$.

Atomic ratios for $7s$ and $6p_{1/2}$ are: $\frac{b_N(7s)}{b_N(6p_{1/2})} = 3.52$ and $\frac{b_M(7s)}{b_M(6p_{1/2})} = 3.30$, while for the H-like ion we had the values of 3.58 and 3.40 respectively.

Situation changes when we include the spin-polarization of the core via RPA corrections. These corrections mix partial waves and the state $6p_{3/2}$ partly acquires and $p_{1/2}$ character. This leads to the significant change of the size and even the sign of the constant $A(6p_{3/2})$. At the same time, this constant becomes very sensitive to the distributions of charge and magnetic moment inside the nucleus.

RPA corrections for the $7s$ and $6p_{1/2}$ states are smaller than for $6p_{3/2}$, but also significant. They lead to

effective mixing of the s and p waves. Because of that, the ratios of the respective coefficients decrease a little, but are still markedly larger than unity:

$$\frac{b_N(7s)}{b_N(6p_{1/2})} = 2.65, \quad \frac{b_M(7s)}{b_M(6p_{1/2})} = 2.44.$$

We conclude that in the DHF+RPA approximation, the anomaly for the $7s$ state is still significantly stronger, than for $6p_{1/2}$ state. The anomaly for the $6p_{3/2}$, on the contrary, becomes the largest. In this work we did not take into account more correlation corrections, but RPA contribution influences the anomaly more strongly than others and changes its behavior. When all corrections are taken into account our method usually gives an accuracy of few percent for neutral atoms, as it was done in [14].

Table 2. Compilation of the fitting parameters for HFS of neutral Tl atom: A_0 is HFS constant for point-like nucleus, δ and ε are the nuclear charge and magnetization distribution corrections parametrized by b_N and b_M coefficients respectively. We use g factor $g_I = 3.27640$. Corrections δ and ε for ^{203}Tl are calculated for $R_N = R_M = 0.1306 \times 10^{-3}$ au.

Calculations are done within DHF and DHF+RPA approximations.

	DHF			DHF+RPA		
	$6p_{1/2}$	$7s$	$6p_{3/2}$	$6p_{1/2}$	$7s$	$6p_{3/2}$
$A_0(\text{GHz})$	18.130	8.855	1.289	22.684	12.120	-2.4711
b_N	0.1054	0.3708	0	0.1400	0.3714	0.5769
$\delta(^{203}\text{Tl})$	0.0302	0.1064	0	0.0402	0.1066	0.1656
b_M	0.0195	0.0643	0	0.0254	0.0619	0.0933
$\varepsilon(^{203}\text{Tl})$	0.0056	0.0185	0	0.0073	0.0178	0.0268

Using the experimentally measured value of the HFS anomaly (10) for the ground state $6p_{1/2}$ of the thallium two stable isotopes $^{205}\Delta^{203}(6p_{1/2}) = -1.036(3) \times 10^{-4}$ [6] and the ratios (21) calculated here, we can obtain corresponding value for the $7s$ state within $R_M=R_N$ approximation: $^{205}\Delta^{203}(7s) = -2.6 \times 10^{-4}$. This value is significantly lower, than experimental value of $-4.7(1.5) \times 10^{-4}$ obtained in Ref. [7].

4. CONCLUSIONS

In this work, we propose a method for calculation hyperfine structure constants of many-electron atoms as functions of nuclear charge and magnetic radii R_N and R_M . The HFS anomaly in this method can be parametrized by b_N and b_M coefficients. If HFS anomaly is known from the experiment, then we can use coefficients b_N and b_M to determine differences between these radii. Alternatively, we can use these coefficients to improve the accuracy for nuclear g factors of the short-lived isotopes, obtained from the ratios of the HFS constants. We tested this method by calculating the HFS constants of H-like thallium ion and obtained fairly good agreement with the analytical expressions from Refs. [5, 11]. Then we made the calculations for neutral thallium atom described as a one-electron system. In the Dirac-Hartree-Fock approximation, the ratios between hyperfine anomalies of s and $p_{1/2}$ states of the neutral Tl atom and respective H-like ion are the same. However, when we include spin-polarization of the core

via RPA corrections, only the hyperfine anomaly for the $7s$ state remains stable. The ratios between $7s$ and $6p_{1/2}$ states change by roughly 30%, and the anomaly for the $6p_{3/2}$ state becomes very large. We conclude that for the precision measurements of g factors it is preferable to use the hyperfine constants for s states, while the $p_{3/2}$ states are least useful.

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