Magnetic Moment Distributions in Tl Nuclei

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The distributions of nuclear magnetic moment gives rise to small changes of the hyperfine structure. This work presents quantitative calculations linking observed "hyperfine anomalies" in Tl to changes in the magnetic moment distribution, with consistent results for the $6p$ and $7s$ states. It is found that the change in magnetic radius between the two stable isotopes is about twice as large as the corresponding change for the charge distribution. This may have some implications for the "Schiff moments" used in the interpretation of experiments searching for P and T violating effects.

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The distribution of protons, neutrons, and magnetic moment vary between different isotopes of an element. The changing *charge* distribution leads to the field isotope shift, studied extensively, e.g., in chains of radioactive Tl isotopes [1,2]. The distribution of charge and of nuclear *magnetic moment* both affect the hyperfine structures (hfs), and measurements of the resulting "hyperfine anomalies" have been performed since the early days of radio-frequency spectroscopy for several elements including Tl [3,4]. However, a quantitative link between observed anomalies and changes in the magnetic moment distribution had to await the development of appropriate methods to treat heavy many-electron systems. This work is the first application of atomic many-body techniques to hfs anomalies. The results provide a calibration for nuclear structure calculations, needed, e.g., to account for the effect of the changing *neutron* distribution in studies of isotope dependent atomic parity nonconservation (PNC) [5]. The atomic wave functions, obtained in the relativistic coupled-cluster approach [6], lead to accurate results for several properties, giving hope that it will be possible to improve the theoretical PNC result for Tl, as needed in view of recent experimental developments [7].

The magnetic moment $\mu = g_s \mathbf{S}_n + g_l \mathbf{L}_n$ of a nucleus (with $I \neq 0$) gives rise to a hyperfine interaction, which can be described by the Hamiltonian

$$h_p^{\mathrm{hfs}} = \hat{\mathbf{r}} \cdot \frac{\boldsymbol{\mu} \times \boldsymbol{\alpha}}{r^2} \tag{1}$$

for a point magnetic dipole. Through comparison with independently determined nuclear magnetic moments [8], the magnetic hfs can be used to test the quality of the wave functions used in the extraction of other properties.

The form of the hyperfine interaction for a general distribution $\rho_m(R)$ of the magnetic moment [normalized so that $\int \rho_m(R) R^2\, dR = 1$] can be obtained from that for a nuclear magnetic moment distributed on a shell with radius R [9]. For the part arising from an *orbital* angular momentum, the interaction is obtained by replacing $1/r^2$ in Eq. (1) by r/R^3 inside the nucleus, whereas the part $\mathbf{S}_n/r^2$ of the interaction due to a spin magnetic moment is replaced inside the nucleus by the "spin asymmetry operator" $\sqrt{10}\,(\mathbf{S}_n \mathbf{C}_n^2)^1 r/R^3$. A change in the magnetic moment distribution then changes the ratio A/g_I between the nuclear $g_I = \mu/I$ factor and the observed hyperfine constant, A. To find the size of this "Bohr-Weisskopf" [9] effect we integrate the difference between the expression (1) for a point nucleus and the interaction for a magnetic moment distributed on a shell. This gives a perturbation

$$h_{\mathrm{BW}}^{\mathrm{hfs}} = \int \left\{ \eta(R - r) \left[g_s \left(\frac{\mathbf{S}_n}{r^2} - \sqrt{10}\,(\mathbf{S}_n \mathbf{C}_n^2)^1 \frac{r}{R^3} \right) + g_L \mathbf{L}_n \left(\frac{1}{r^2} - \frac{r}{R^3} \right) \right] \times \boldsymbol{\alpha} \right\} \rho_m(R) R^2\, dR\,, \tag{2}$$

where the step function $\eta(R - r)$ is 1 for $r \leq R$ and 0 elsewhere.

The relative change in the hfs constant due to the distributed nuclear moment for a state Ψ is given by

$$\epsilon_{\mathrm{BW}} = \langle \Psi | h_{\mathrm{BW}}^{\mathrm{hfs}} | \Psi \rangle / \langle \Psi | h_p^{\mathrm{hfs}} | \Psi \rangle\,.$$

With the exception of recent experimental investigations of the ground state hfs in heavy hydrogenlike ions [10], this effect cannot be observed for a single isotope, since the electronic factor for the hyperfine structure is not known with sufficient accuracy. What can be observed, however, is a *shift* in the ratio between hyperfine constants and nuclear g factors for two isotopes,

$$\frac{A_1 g_2}{A_2 g_1} - 1 \equiv \Delta_{12}\,.$$

The effect of an extended charge distribution on the hfs gives the "Breit-Rosenthal(-Crawford-Schawlow)" correction, ϵ_{BR}, which was estimated in early works [11–13] by analytical solutions for particular nuclear distributions. In this work, this correction is instead parametrized by noting that, as known from studies of isotope shifts, the varying charge distribution results in a perturbation proportional to $\delta\langle r_c^2 \rangle$ or, rather, to

$$\lambda_c = \kappa_c \delta \langle r_c^2 \rangle\,, \tag{3}$$

where the factor κ_c accounts for higher moments of the nuclear charge. It is slightly model dependent, but

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for a uniform charge distribution κ_c is found to have a value of about 0.94 for $Z = 81$ [14]. This perturbation modifies the wave function and thereby the hfs. By performing Dirac-Fock calculations for several nuclear charge distributions, ρ_c, and comparing the results to a reference nucleus with nuclear charge and magnetic moment distributions, $\rho_{c,0}$ and ρ_m, respectively (both with $R_N \approx 7$ fm), we found

$$A(\rho_c, \rho_m) \approx A(\rho_{c,0}, \rho_m)(1 - f\lambda_c), \quad (4)$$

where the factors f were found to be $f(7s) = 15.8 \times 10^{-4}/\text{fm}^2$ and $f(6p_{1/2}) = 4.14 \times 10^{-4}/\text{fm}^2$.

To investigate the effect of the distribution of the magnetic moment, we note first that the one-electron hyperfine matrix element involves the cross product P_{od} of the radial parts of the upper and lower components, of the relativistic wave function which for small r can be expressed as $P_{od} = a_1 r + a_3 r^3 + a_5 r^5 + \cdots$ for $j = 1/2$ state (a_k vanishes for $k < 2j$). Combining P_{od} with the perturbation h_{BW} in Eq. (2) we find that the Bohr-Weisskopf contribution to the hfs anomaly can be expressed as

$$\Delta_{\text{BW}} = b_2 \delta\langle r_m^2\rangle + b_4 \delta\langle r_m^4\rangle + \cdots . \quad (5)$$

The coefficients b_{2n} in Eq. (5) get different contributions for the various terms in $h_{\text{BW}}^{\text{hfs}}$, giving

$$b_{2n} = C_S(b_{2ns} + \zeta b_{2nd}) + C_L b_{2nl} \equiv b_{2ns} d_{2n}, \quad (6)$$

where C_S and C_L are the "fractional contributions of spin and orbital moment to the magnetic moment" ($C_S + C_L = 1$) [15]. The factor ζ for the spin-asymmetry term can be evaluated using standard tensor techniques, and its values for single-particle states can be found, e.g., in Refs. [9,15,16]. The value for the coefficient d_{2n} in Eq. (6) can be simplified by using the relations [17] $b_{2n,l} = 3b_{2n,s}/(2n + 3) = b_{2n,s} - b_{2n,d}$ giving

$$d_{2n} = C_S\left(1 + \frac{2n}{2n + 3}\zeta\right) + \frac{3}{2n + 3}(1 - C_S). \quad (7)$$

The coefficients b_{2s}, b_{4s}, and b_{6s} have been evaluated using numerical Dirac-Fock orbitals, obtained in a homogeneous charge density with $R_N = 1.2A^{1/3}$ fm, for $A = 203$, gave $b_{2s}(6p_{1/2}) = 2.26 \times 10^{-4}/\text{fm}^2$ and $b_{2s}(7s) = 7.95 \times 10^{-4}/\text{fm}^2$. To a good approximation, these coefficients depend only on the angular momentum, but are independent of the orbital energy, and thus hold also for excited orbitals. The correction due to higher moments of the distribution are essentially the same: in both cases $b_{4s}/b_{2s} \approx -0.0032/\text{fm}^2$ and $b_{6s}/b_{2s} \approx 8.8 \times 10^{-6}/\text{fm}^4$.

We now rewrite the expression (5) for the Bohr-Weisskopf contribution to the hyperfine anomaly as

$$\Delta_{\text{BW}} = b_{2s} d_2 \delta\langle r^2\rangle + b_{4s} d_4 \delta\langle r^4\rangle + \cdots = b_{2s}\lambda_m, \quad (8)$$

where we used d_{2n} from Eqs. (6) and (7) and introduced

$$\lambda_m = \delta\langle r^2\rangle\left(d_2 + \frac{b_{4s}d_4}{b_{2s}}\frac{\delta\langle r^4\rangle}{\delta\langle r^2\rangle} + \cdots\right). \quad (9)$$

The values for b_{2s} can be compared to the Dirac-Fock values for the factor f in Eq. (4) for the sensitivities to the changes in charge distribution, and we find the ratios $f(7s)/b_{2s}(7s) \approx 2.0$ and $f(6p_{1/2})/b_{2s}(6p_{1/2}) \approx 1.8$. [The relative difference between these numbers is essentially $2mc^2/(3Ze^2/2R_N)$ arising from the different boundary conditions for the upper and lower components of the orbitals.] The effect of the changes in charge and magnetic dipole distributions on the hfs can thus be combined into a single parameter $\lambda_{c,m}$:

$$\lambda_{c,m} = \lambda_m + 1.91(1)\lambda_c . \quad (10)$$

We note here that a similar parametrization can be applied to the results presented by Finkbeiner, Fricke, and Kühl [18] for the ground states hfs in H-like Bi ($Z = 83$). Their data can be summarized as $\delta\nu \approx \delta\nu_0[1 - (8.2 \times 10^{-4}/\text{fm}^2)\lambda_{c,m}]$ and are consistent with $\lambda_{c,m}$ from Eq. (10). If the reference nucleus is chosen as their Fermi nucleus with $c = 6.65$ fm, $t = 2.28$ fm, and a point magnetic dipole, then $\delta\nu_0 = 4.189 \times 10^{-4}$ cm^{-1}. The corrections to $\delta\nu_0$ depend on details of the nuclear structure.

The parameter $\lambda_{c,m}$ will be useful for a many-electron system like Tl only if the ratio f/b_{2s} holds also for many-body corrections—as it can, however, be expected to do, since changed distributions of charge and magnetic moments alike affect mainly matrix elements between $j = 1/2$ states. The relation was confirmed in evaluations of "RPA" (random phase approximation) effects (following the procedure described, e.g., in Ref. [19]), which gave large contributions to the hyperfine anomalies for all states, whereas the ratio f/b_{2s} was found to be essentially constant (even for the strongly perturbed $6p_{3/2}$ state). We then expect it to be unchanged also by correlation effects and evaluate only the coefficient for $\lambda_{c,m}$.

Tables I and II present the results for hyperfine constants A and anomalies in the form $\Delta/\lambda_{c,m}$ obtained for the $6p$ and $7s$ states of Tl using the relativistic "coupled-cluster" approach following the procedures applied earlier [20,21] to calculations for Ba^+ and Yb^+. The calculations presented here include single and double excitations with respect to the unperturbed core of Tl^+ involving excitations of the core orbitals $4f$, $5p$, $5d$, and $6s$ up to a maximum angular momentum of 6 for the core-valence pair excitations and a maximum angular momentum of 4 for the core-core excitations. The resulting wave functions were used to evaluate also other properties, for which final results are shown in Table III and compared to experimental data, where available. A detailed description of the terms included can be found in Ref. [22]. Although the results obtained here seem to be the best calculated values to date, an even more detailed treatment is required for some properties, probably involving either three-particle excitations or a more general model space as a starting point.

The comparison for the isotope shifts shown in Table III is somewhat hampered by the uncertainty in the calculated specific mass shift constant [23], and thus the

TABLE I. Comparison of calculated hyperfine structure constants A for ^{205}Tl with results obtained in other calculations and with experimental values (GHz).

	$A(6p_{1/2})$	$A(6p_{3/2})$	$A(7s)$
DF	17.68	1.304	7.78
BO	2.24	0.21	2.22
RPA	1.98	−1.64	2.88
Corr	−1.04	0.39	−0.20
Total	20.86	0.256	12.67
+ inner RPA[a]	21.43	0.317	12.92
+ finite size[b]	21.3	0.339	12.76
Other calculations			
SDCF[c]	18.73	1.381	
MCDF[d]	20.32	1.485	
g Hartree[e]	20.89	0.895	
BO + RPA[f]	24.06[g]	−1.885[g]	13.06[g]
	21.77[h]	−1.919[h]	12.47[h]
+ "int corr"[i]	21.3	0.600	12.56
Experiment	21.3108[j]	0.2650[k]	12.2972[c]

[a]This line gives an estimate of the RPA correction from core orbitals not included in the coupled-cluster calculation.
[b]The finite size corrections were estimated using the factors from Table II.
[c]Hermann *et al.*, Z. Phys. D **28**, 127 (1993).
[d]The MCDF hfs values include increments from separate MDCF results, obtained by Grexa *et al.*, Phys. Rev. A **38**, 1263 (1988).
[e]Millack, Z. Phys. D **8**, 119 (1988).
[f]Brueckner orbitals + RPA corrections.
[g]Hartley and Mårtensson-Pendrill, Z. Phys. D **15**, 309 (1991).
[h]Dzuba *et al.*, J. Phys. B **8**, 597 (1985).
[i]BO + RPA + "internal correlation," Dzuba *et al.*, J. Phys. B **20**, 1399 (1987); Phys. Lett. A **131**, 461 (1988).
[j]Lurio and Prodell, Ref. [3].
[k]Gould, Ref. [4].

values extracted from accurate laser spectroscopic experiments cannot yet compete with the muonic determination for the stable Tl isotopes [24,25]. Nevertheless, the calculated F factors do provide a scale for extracting $\delta\langle r_c^2\rangle$ from the isotope shift measurements on chains of radioactive Tl isotopes [1,2].

In Table II, we have given the correction (expressed in terms of b_{2s}) due to distributed nuclear charge and magnetic moment for the various contributions to the hfs (shown in Table I) of the $6p$ and $7s$ states. These corrections reflect the interplay between admixtures of various angular momenta into the wave function.

These results can be combined with experimental data for $^{203:205}$Tl. As shown in Table II, this leads to consistent values for the change $^{203:205}\lambda_{c,m}$ between the two stable Tl isotopes, even for the strongly perturbed $6p_{3/2}$ state. In the analysis below, we use $^{203:205}\lambda_{c,m} = 0.42$ fm^2 from the results for $6p_{1/2}$, which should be the most accurate value. The contribution from λ_c is obtained by combining the muonic value [24,25] $^{203:205}\delta\langle r_c^2\rangle = 0.115(3)$ fm^2 with Eqs. (3) and (10), giving $\lambda_m = \lambda_{c,m} - 0.21(1)$ fm$^2 \approx 0.21(1)$ fm^2, so the Tl hyperfine anomaly gets about equal contribution from the Bohr-Weisskopf effect and from the Breit-Rosenthal correction.

TABLE II. Hyperfine anomalies: The theoretical values give the relative effect of the magnetic moment distributions on the various terms shown in Table I and are given in terms of b_{2s} factors, i.e., as $\Delta/\lambda_{c,m}$. The experimental hyperfine anomalies were obtained using the magnetic moments [8] $^{203}\mu = 1.622\,257\,87\mu_N$ and $^{205}\mu = 1.638\,314\,61\mu_N$. The $\lambda_{c,m}$ values shown in the last line were extracted by combining theoretical and experimental values, but the error bars do not reflect the theoretical uncertainty.

	$6p_{1/2}$	$6p_{3/2}$	$7s$
$\Delta/\lambda_{c,m}(10^{-4}/\text{fm}^2)$			
DF(BO)	−2.26	0	−7.95
RPA	−4.89	−5.02	−5.86
Corr	−4.12	−4.24	−1.63
Total	−2.48	43.0	−7.62
Experiment			
$\Delta(10^{-4})$	−1.04[a]	16.26[b]	−3.4(18)[c]
$\lambda_{c,m}$ (fm^2)	0.42	0.38	0.45(24)

[a]Lurio and Prodell, Ref. [3].
[b]Gould, Ref. [4].
[c]Hermann *et al.*, Z. Phys. D **28**, 127 (1993).

For the case where the magnetic moment is mainly of spin character, as for ^{203}Tl and ^{205}Tl which both have $I = 1/2$ and a valence proton described by $3s_{1/2}$ in the shell model, we find that $d_2 \approx 1$ in Eq. (9) for λ_m. This leads to $^{203:205}\delta\langle r_m^2\rangle = 0.26(2)$ fm^2. The error bar includes an uncertainty in the calculated values which should be no worse than 1%. It also includes an uncertainty due to the correction of higher moments of the magnetic moment

TABLE III. Theoretical results obtained for other properties and comparison with experimental results, where available.

	Theory	Experiment
Field isotope shift constants (GHz/fm^2)		
$F(6p_{1/2})$	10.13	11.3(6)[a]
$F(6p_{3/2})$	11.43	12.1(6)[a]
$F(7s)$	−4.22	−3.87(3)[a]
Reduced transition matrix elements		
$E1$ ($7s \rightarrow 6p_{1/2}$) (ea_0)	1.78(2)[b]	1.82(5)[c]
$E1$ ($7s \rightarrow 6p_{3/2}$) (ea_0)	3.31(8)[b]	3.27(7)[c]
$E2$ ($6p_{1/2} \rightarrow 6p_{23}$) ($e^2a_0^2$)	13.26	13.29(3)[d]

[a]The experimental F values were obtained by combining the experimental level isotope shifts [Grexa *et al.*, Phys. Rev. A **38**, 1263 (1988)], calculated specific mass shift constants [23], and the value $^{203:205}\delta\langle r^2\rangle = 0.115(3)$ fm^2 obtained in muonic experiments [24]. The uncertainty reflects the uncertainty in the estimate of the specific mass shift.
[b]The values are obtained from a combination of results obtained in the length and velocity form, and the uncertainty is taken as the correcton to the value in the "length" form.
[c]Gallagher and Lurio, Phys. Rev. **136**, A87 (1964).
[d]Vetter *et al.*, Ref. [7].

distribution, but not uncertainties in the fraction $C_S \approx 1$ in Eq. (7) of spin magnetic moment nor in the spin asymmetry parameter $\zeta \approx 0$, which can change slightly due to admixture of other nuclear states.

Limited nuclear structure calculations were performed by Tomaselli, Herold, and Grünbaum [26], who got qualitative agreement with experimental data for the magnetic moments. They also evaluated the muonic hyperfine structure and found a ratio to the magnetic moment which was larger for ^{205}Tl than for ^{203}Tl, which would seem to indicate that the magnetic moment distribution of ^{203}Tl was more extended, whereas the atomic data show that both the charge and magnetic radius are larger for ^{205}Tl, as one would naively expect.

By combining modern atomic structure calculations with experimental hyperfine anomalies obtained in the 1950s we have found that the change in magnetic radius between ^{203}Tl and ^{205}Tl is more than twice as large as the change in charge radius. This may have implications for "Schiff moments" which describe the difference between the distributions of nuclear charge and a possible P and T violating nuclear *electric* dipole moment [27–29]. In fact, one of the lowest upper limits for a nuclear Schiff moment is that obtained for ^{205}Tl from experiments on TlF [30]. It is hoped that the analysis of old results presented here will provide calibration and stimulation for the improved nuclear structure calculations for Tl.

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