Supplementary Materials

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The magnetic dipole moments (μ) of ^{209}Tl^{g} ($N = 128$) and ^{207}Tl^{m} ($N = 126$) have been measured for the first time using the in-source laser resonance-ionization spectroscopy technique with the Laser Ion Source and Trap (LIST) at ISOLDE (CERN). The application of the LIST suppresses the usually overwhelming background of the isobaric francium isotopes and allows access to heavy thallium isotopes with $A \geqslant 207$. The self-consistent theory of finite Fermi systems based on the energy density functional by Fayans *et al*. well describes the N dependence of μ for $1/2^+$ thallium ground states, as well as μ for the $11/2^-$ isomeric states in europium, gold and thallium isotopes. The inclusion of particle-vibration coupling leads to a better agreement between the theory and experiment for μ(Tl^{g}, $I^{\pi} = 1/2^+$). It is shown that beyond mean-field contributions to μ cannot be neglected at least for thallium isotopes with $I^{\pi} = 1/2^+$.

1. Suppression of ^{208}Fr in the LIST mode

Figure 1 depicts the γ-ray spectrum for ^{208}Tl laser scan at the maximum of the hfs. Comparable amounts of the ^{208}Tl and ^{208}Fr decay are seen. At the same time, the yield of ^{208}Fr would be approximately 3-4 orders of magnitude higher than that of ^{208}Tl if the LIST mode of operation was not used. Note, that ^{208}Tl was suppressed by a factor of ~20 in this spectrum due to the LIST operation.

2. Electric field gradient for the $6d\ ^2D_{3/2}$ state of Tl

The value of the electric field gradient (V) for the $6d\ ^2D_{3/2}$ state of thallium was calculated employing the

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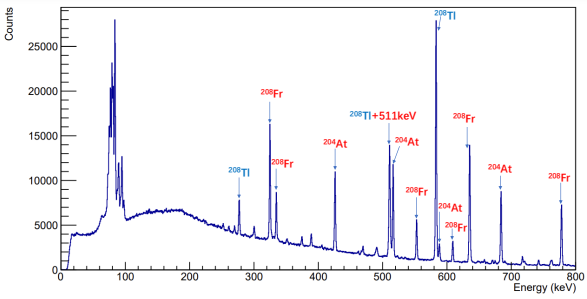

Figure 1: A part of γ-ray spectrum (up to 800 keV) measured at the maximum of hfs for ^{208}Tl. The main peaks coming from ^{208}Tl decay, together with the decay of the isobaric contamination, ^{208}Fr, and its daughter product, ^{204}At, are labelled. ^{204}At is produced via α decay of ^{208}Fr, which has a half-life of 59.1 s and α branching ratio of 89%. The labelled **^{204}At** peaks are from the β-delayed γ-rays of ^{204}At. Peaks at 583 keV and 2614.5 keV were used to construct the hfs for ^{208}Tl. The measurement was performed under the LIST mode.

relativistic coupled cluster theory and the Dirac-Coulomb Hamiltonian. In the correlation calculation within the coupled cluster approach with single, double and perturbative triple cluster amplitudes CCSD(T) [1, 2] all electrons were correlated and the uncontracted Dyall's all-electron quadruple-zeta AE4Z [3, 4] basis set augmented by several diffuse functions was used as in Ref. [5]. To take into account higher-order correlation effects we calculated the difference between the EFG values obtained within the coupled cluster with single, double, triple and perturbative quadruple amplitudes CCSDT(Q) [6, 7, 8] method and the CCSD(T) one. Here 21 electrons were correlated and the AE3Z [3, 4] basis set augmented by diffuse functions [5] was employed. The accuracy of such scheme was analyzed previously for other properties and found to give precise results [5, 9, 10]. The final result of calculations gives V = 41.2(23) MHz/b

In our previous papers on IS in thallium [11, 12] we used systematics of a and b constants for $nd\,^2D_{3/2}$ states (n = 3-5) of the homologs of Tl (Al, Ga and In) and one-electron approximation [13] to estimate the EFG($6d\,^2D_{3/2}$): V(one-electron)=15 MHz/b. Although the correct calculations give EFG more than two times larger than the crude one-electron estimations, the conclusion about the possibility to neglect $b(6d\,^2D_{3/2})$ made in [11] remains valid.

3. Hyperfine anomaly

It was shown that for some thallium isotopes RHFA can reach 1% [14], that is, it can be comparable with our experimental uncertainties. Thus, we cannot neglect this correction. There are two independent contributions in RHFA, connected with the change of the charge (Breit-Rosenthal, BR correction) and magnetization (Bohr-Weisskopf, BW correction) distribution in the nucleus, respectively [15, 16, 17, 18]. However, according to the calculation of Ref. [19], for Tl isotopes in the range $A = 203-209$, the BR correction proves to be very small ($< 2 \cdot 10^{-4}$) and it can be neglected in comparison with the BW correction and experimental uncertainties. Correspondingly, only the BW anomaly will be considered and the index "BW" will be omitted.

The RHFA for ^{207,209}Tl^g were estimated in accordance with the procedure outlined in Refs. [20, 14, 21, 22]. Namely, the differential HFA (DHFA), ${}^{A_1}_{6P_{1/2}}\Delta^{A_2}_{7S_{1/2}}$, is introduced which relates the change in the ratio of the magnetic hfs constants for different atomic states ($6P_{1/2}$ and $7S_{1/2}$ in our case) to the difference of the corresponding RHFA values:

$$\rho^{A}_{6P_{1/2},7S_{1/2}} \equiv \frac{a_A(6P_{1/2})}{a_A(7S_{1/2})} \tag{1}$$

$$ {}^{205}_{6P_{1/2}}\Delta^{A}_{7S_{1/2}} \equiv \frac{\rho^{205}_{6P_{1/2},7S_{1/2}}}{\rho^{A}_{6P_{1/2},7S_{1/2}}} - 1 \approx \approx {}^{205}\Delta^{A}(6P_{1/2}) - {}^{205}\Delta^{A}(7S_{1/2}) \tag{2}$$

The BW contribution to the magnetic hyperfine structure constant can be factorized into the electronic part and the nuclear-magnetization dependent part [9, 23]. The electronic part is independent of A, which means that the ratio

$$\eta_{7S_{1/2},6P_{1/2}} \equiv \frac{{}^{205}\Delta^{A}(7S_{1/2})}{{}^{205}\Delta^{A}(6P_{1/2})} \tag{3}$$

is determined solely by the electronic structure and RHFA can be calculated using the experimental value of DHFA:

$${}^{205}\Delta^{A}(6P_{1/2}) = \frac{{}^{A_1}_{6P_{1/2}}\Delta^{A_2}_{7S_{1/2}}}{1-\eta_{7S_{1/2},6P_{1/2}}} \tag{4}$$

The factor η was calculated within the coupled cluster method with single, double and perturbative triple cluster amplitudes based on the Dirac-Coulomb Hamiltonian and correlating all electrons [23]: $\eta_{7S_{1/2},6P_{1/2}} = 3.54(14)$ (with the uncertainty covering electronic wave function uncertainty and slight model dependence of η). Using $a_{207g}(7S_{1/2}) =$ 14070(7) MHz [24], $a_{207g}(6P_{1/2}) = 24390(105)$ MHz (present work, see Table I), $a_{205}(7S_{1/2}) = 12296.09(70)$ MHz [25], $a_{205}(7P_{1/2}) = 21310.835(5)$ MHz [26] and calculated η factor, one obtains from Eqs. (2,3,4): $|{}^{205}\Delta^{207}(6P_{1/2})| < 2 \cdot 10^{-3}$

Unfortunately, there are no data on $a_{209}(7S_{1/2})$ and $a_{207m}(7S_{1/2})$ and we cannot estimate the RHFA for ^{209}Tl and ^{207}Tl^m exclusively from experimental information. However, systematic inspection of the RHFA (see [27, 28]) shows that the RHFA value correlates with the value of the magnetic moment at least for isotopes with the same leading nuclear configuration. This is the case for ^{203,205,207,209}Tl where ground states are predominately of $\pi s_{1/2}$ configuration. Our measurements show that $\mu(^{203}\text{Tl}) < \mu(^{209}\text{Tl}) < \mu(^{207}\text{Tl}^g)$. Correlation of magnetic moment and RHFA means that $|{}^{205}\Delta^{209}(6P_{1/2})| < |{}^{205}\Delta^{207}(6P_{1/2})|$. Thus,

estimation of $^{205}\Delta^{207}(6P_{1/2})$ is obviously valid for ^{209}Tl, i. e. we conservatively adopt $|^{205}\Delta^{209}(6P_{1/2})| < 2 \cdot 10^{-3}$.

Atomic-nuclear factorization enables one to present the RHFA for $^{207}\mathrm{Tl}^m$ in the following form [19, 21]:

$$\begin{aligned} &^{205}\Delta^{207m}(6P_{1/2}) = \\ &= b_M^{6P}(R/\lambda\!\!\!^{-}_C)^{\kappa}[d_{nuc}(\pi s_{1/2}) - d_{nuc}(\pi h_{11/2})] \end{aligned} \tag{5}$$

where $\kappa = 2\sqrt{1-(\alpha Z)^2}-1$, $\lambda\!\!\!^{-}_C$ is the Compton wavelength of the electron, α is the fine-structure constant, and the dimensionless parameter b_M depends on the electronic wave function of the atom. A similar formula is valid for $\pi h_{9/2}$ states. Thus, RHFAs for $\pi h_{11/2}$ and $\pi h_{9/2}$ single-particle states in thallium are connected by the relation:

$$\begin{aligned} &^{205}\Delta^{207m(I=11/2)}(6P_{1/2}) = \\ &= \frac{[d_{nuc}(\pi s_{1/2}) - d_{nuc}(\pi h_{11/2})]}{[d_{nuc}(\pi s_{1/2}) - d_{nuc}(\pi h_{9/2})]}\times \\ &\times\, ^{205}\Delta^{(I=9/2)}(6P_{1/2}) \end{aligned} \tag{6}$$

Thus, with the RHFA for the $\pi h_{9/2}$ state deduced from the experimental data ($^{205}\Delta^{(I=9/2)}(6P_{1/2}) = -0.007(2)$ [14]), one can calculate the RHFA for the $\pi h_{11/2}$ state in $^{207}\mathrm{Tl}^m$, provided the corresponding nuclear factors are determined. In the single-particle approximation this factor can be calculated by the formulas from Refs. [17, 18].

It follows from Eq. (5) that the DHFA $^{205}_{6P_{1/2}}\Delta^{A(I=9/2)}_{7S_{1/2}}$ is proportional to $^{205}\Delta^{(I=9/2)}(6P_{1/2})$. It was shown in [14] that experimental value of this DHFA differs from the theoretical one by 27%. We conservatively relate all this difference to the uncertainty of the theoretical nuclear factor $[d_{nuc}(\pi s_{1/2}) - d_{nuc}(\pi h_{9/2})]$. Note, that the magnetic moment of the $\pi h_{9/2}$ thallium states is weakly depended on N (it changes by 10% when going from $N = 116$ to $N = 102$ [11, 12]). The magnetic moment of the $\pi h_{11/2}$ state displays the same behavior (changing by 15% for isotopes with different Z near $Z = 82$ and $N = 82 - 126$) [21]. The stability of the high-spin state magnetic moments in the lead region confirms the predominantly single-particle nature of these states, that is, the applicability of the single-particle approximation for the nuclear-factor calculation and supports the estimation of the upper limit of the theoretical nuclear-factor uncertainty. Keeping in mind the similar behavior of the magnetic moment for $\pi h_{9/2}$ and $\pi h_{11/2}$ states, we ascribe the same 27% uncertainty to the nuclear factor $[d_{nuc}(\pi s_{1/2}) - d_{nuc}(\pi h_{11/2})]$.

Taking into account uncertainties of the experimental RHFA value, $^{205}\Delta^{(I=9/2)}(6P_{1/2})$, and nuclear factors, one obtains from Eq. (6): $^{205}\Delta^{207m}(6P_{1/2}) = -0.0033(16)$.

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