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Excitation energies, hyperfine constants, *E*1, *E*2, and *M*1 transition rates, and lifetimes of $6s^2nl$ states in Tl I and Pb II

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Energies of $6s^2np_j$ ($n=6$–9), $6s^2ns_{1/2}$ ($n=7$–9), $6s^2nd_j$ ($n=6$–8), and $6s^2nf_{5/2}$ ($n=5$–6) states in Tl I and Pb II are obtained using relativistic many-body perturbation theory. Reduced matrix elements, oscillator strengths, transition rates, and lifetimes are determined for the 72 possible $6s^2nl_j$-$6s^2n'l'_{j'}$ electric-dipole transitions. Electric-quadrupole and magnetic-dipole matrix elements are evaluated to obtain $6s^2np_{3/2}$-$6s^2mp_{1/2}$ ($n,m=6,7$) transition rates. Hyperfine constants *A* are evaluated for $6s^2np_j$ ($n=6$–9), $6s^2ns_{1/2}$ ($n=7$–9), and $6s^2nd_j$ ($n=6$–8) states in ^{205}Tl. First-, second-, third-, and all-order corrections to the energies and matrix elements and first- and second-order Breit corrections to energies are calculated. In our implementation of the all-order method, single and double excitations of Dirac-Fock wave functions are included to all orders in perturbation theory. These calculations provide a theoretical benchmark for comparison with experiment and theory.

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I. INTRODUCTION

This work continues earlier relativistic many-body perturbation theory (RMBPT) studies of energy levels of ions with one valence electron outside of a closed core [1–7]. We consider the three-electron system $[\mathrm{Nd}]5s^25p^65d^{10}6s^2nl$ in Tl I and Pb II as a one-electron *nl* system with $[\mathrm{Nd}]5s^25p^65d^{10}6s^2$ core. The ground-state energy of thallium $[\mathrm{Nd}]5s^25p^65d^{10}6s^26p$, treated as a one-electron system, was calculated by Dzuba *et al.* [8] using perturbation theory in a screened Coulomb interaction (PTSCI), by Blundell *et al.* [9] using third-order many-body perturbation theory (MBPT), and by Liu and Kelly [10] using the coupled-cluster (CC) approach. Second-order MBPT energies of thallium were evaluated for the ground and excited states ($6s^26p$, $6s^27s$, and $6s^27p$) by Hartley and Martensson-Pendrill [11]. Thallium was treated as a three-particle system in Refs. [12–15] to evaluate the Tl I ionization potential and the first few energy levels, $6s^26p$, $6s^26d$, and $6s^27s$. The second-order relativistic MBPT was used by Johnson *et al.* [12] and a combined second-order MBPT plus configuration-interaction (CI) method was employed in Refs. [13–15]. In the present paper, the energies of $6s^2np_j$ ($n=6$–9), $6s^2ns_{1/2}$ ($n=7$–9), $6s^2nd_j$ ($n=6$–8), and $6s^2nf_{5/2}$ ($n=5$–6) states in Tl I and Pb II are obtained using the third-order RMBPT and the SD all-order method, in which single and double (SD) excitations of the Dirac-Fock (DF) wave functions are summed to all orders.

A relativistic semiempirical method including exchange was employed by Migdalek [16] to calculate the oscillator strengths in Tl I for $6s^26p_j$-$6s^2ns_{1/2}$, $6s^27p_j$-$6s^2ns_{1/2}$, $6s^26p_j$-$6s^2nd_j$, and $6s^27s_{1/2}$-$6s^2np_j$ transitions. Oscillator strengths determined from single-configuration relativistic Hartree-Fock (RHF) calculations were reported by Migdalek and Baylis [17] for the lowest $6s^26p_j$-$6s^27s_{1/2}$ and $6s^26p_j$-$6s^26d_{j'}$ transitions. A numerical Coulomb approximation (NCA) was used by Lindgärd *et al.* [18] to calculate the lifetimes of $7s_{1/2}$, $8s_{1/2}$, $9s_{1/2}$, $6d_j$, $7d_j$, and $8d_j$ states. Correlation corrections to electric-dipole (*E*1) matrix elements in Tl I were performed in Refs. [11,13,15]. Radial integrals for allowed *E*1 $6p_j$-$ns_{1/2}$ and $6p_j$-$6d_{j'}$ transitions were calculated by Dzuba *et al.* [13] taking into account the second-order diagram contributions. Approximate Brueckner orbitals were used in [11] to evaluate reduced electric-dipole matrix elements for $6p_j$-$ns_{1/2}$ transitions. Dipole matrix elements for $6p_j$-$7s_{1/2}$, $6p_j$-$6d_{j'}$, $7p_j$-$7s_{1/2}$, and $7p_j$-$6d_{j'}$ transitions obtained by combining MBPT and CI methods were presented by Kozlov *et al.* [15].

Calculations of the Pb II properties including a model potential to represent the core polarization of Pb II were reported by Migdalek [19] and by Alonso-Medina [20]. Oscillator strengths and relative line strengths for the $6\ ^2P_J$–$n\ ^2S_{1/2}$ transitions ($n=7$–11) and for the $7\ ^2P_J$–$n\ ^2S_{1/2}$ transitions ($n=8$–11) were presented in Ref. [19]. Transition probabilities for the $7\ ^2P_J$–$n\ ^2S_{1/2}$ ($n=8$–11), $7\ ^2P_J$–$n\ ^2D_{J'}$ ($n=7,8,10$), $7\ ^2S_{1/2}$–$7\ ^2P_J$, and $6\ ^2D_J$–$n\ ^2F_{J'}$ ($n=5$–7) transitions were calculated by Alonso-Medina [20]. Transition

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probabilities for 190 lines arising from the $6s^2ns\ ^2S_{1/2}$, $6s^2np\ ^2P_J$, $6s^2nd\ ^2D_J$, $6s^2nf\ ^2F_J$, $6s6p^2\ ^2S_{1/2}$, $^{2,4}P_J$, and $^{2,4}D_J$ levels in Pb II were also calculated recently by Colon and Alonso-Medina [21] using the COWAN code. Additionally, theoretical lifetimes were calculated in [21] for those levels for which lifetimes measurements were given in [22]. Those theoretical results included correlation corrections by using scaled radial integrals in the COWAN code.

In this paper, we conduct both MBPT and SD all-order calculations of Tl I and Pb II properties. Such calculations permit one to investigate the convergence of perturbation theory and estimate the theoretical error of predicted data. In the present paper, we evaluate reduced matrix elements, oscillator strengths, transition rates, and lifetimes for the 72 possible nl_j-$n'l'_{j'}$ electric-dipole transitions in Tl I and Pb II. Our results are compared with theoretical results from Ref. [15] and with experimental measurements from Refs. [23–26] in Tl I and Refs. [21,27] in Pb II.

The relativistic Hartree-Fock (HFR) COWAN code was used by Biemout and Quinet [28] to calculate the $6\ ^2P_{1/2}$–$6\ ^2P_{3/2}$ magnetic-dipole and electric-quadrupole transition probabilities in Tl I and Pb II. Magnetic-dipole and electric-quadrupole transition rates for $n\ ^2P_{1/2}$–$n'\ ^2P_{3/2}$ transitions $(n,n'=6,7)$ in Tl were presented by Neuffer and Commins [29], where valence-electron wave functions were generated as numerical solutions of the Dirac equation in a modified Tietz central potential. In present paper, we extend those studies by using relativistic MBPT and SD all-order methods to obtain *ab initio* results with detailed consideration of the correlation effects.

Approximate Brueckner orbitals were used in [11] to evaluate the hyperfine constants for the ground and excited states ($6p_{1/2}$, $6p_{3/2}$, $7s_{1/2}$, and $7p_{1/2}$) in ^{205}Tl. Results for the hyperfine constants for $6p_j$, $7p_j$, $7s_{1/2}$, and $6d_j$ states in ^{205}Tl were obtained by combining MBPT and CI methods by Kozlov *et al.* [15]. In the present paper, we calculate hyperfine constants A for $6s^2np_j$ $(n=6-9)$, $6s^2ns_{1/2}$ $(n=7-9)$, and $6s^2nd_j$ $(n=6-8)$ states in ^{205}Tl using the relativistic MBPT and SD all-order methods. Our SD results are compared with the theoretical results from Ref. [15] and with experimental measurements from Refs. [29–31].

In summary, this work presents both a systematic calculation of various Tl I and Pb II properties and a study of the importance of high-order correlation corrections to those properties. One of the aspects of our work was to study the effectiveness of the all-order method when it is applied to three-particle systems such as Tl or Pb$^+$. The advantage of the all-order method is that it treats correlations in a rather compete way yielding excellent results for the alkali-metal atoms. We note that its implementation is entirely different from the configuration interaction method. The trade-off for the application of the all-order method is that its derivation and formulation is complicated even for the simplest case of the systems with one electron above the closed shells. Moreover, the method is numerically demanding. Our work has established that the all-order method is clearly suitable for accurate calculations of properties of Tl and Pb$^+$. By performing calculations using both the all-order method and third-order MBPT, we were able to study the relative importance of correlation corrections on certain properties (by looking for a significant improvement in results when the all-order method is used) and the cases where the treatment of Tl as a three-particle system may be important (where significant discrepancies still remain in the all-order case). The development of the all-order method which is capable of fully treating Tl or Pb$^+$ as a three-particle system is a difficult task and the initial study of the applicability of the all-order method to Tl as a one-particle system is necessary. We find that the SD all-order method works well for the calculation of the Tl properties even without consideration of the three-particle states. Another important result of this study is that we did not find the poor agreement between third-order energies and experiment that occurs for all heavy alkali-metal atoms. It is known [1,6] that the addition of third-order contributions to the energy for heavy alkali-metal atoms does not bring energies into closer agreement with experiment. We find, however, that this is not the case for Tl I and Pb II, where third-order calculations produce values in reasonably good agreement with experiment and all-order SD calculations, when modified to include all third-order contributions, give even better results.

II. ENERGIES OF Tl I AND Pb II

We start from the "no-pair" Hamiltonian [32]

$$H = H_0 + V_{\mathrm{I}}, \tag{1}$$

where H_0 and V_{I} can be written in a second-quantized form as

$$H_0 = \sum_i \varepsilon_i a_i^\dagger a_i, \tag{2}$$

$$V_I = \frac{1}{2} \sum_{ijkl} g_{ijkl} a_i^\dagger a_j^\dagger a_l a_k, \tag{3}$$

where negative energy (positron) states are excluded from the sums. The quantities ε_i are eigenvalues of the one-electron Dirac-Fock equations with a frozen core and the two-particle matrix element g_{ijkl} is the Coulomb matrix element.

Considering neutral Tl as a system with one valence electron we carry out our calculations starting with the V^{N-1} DF potential $[\mathrm{Nd}]5s^25p^65d^{10}6s^2$. There are a number of advantages associated with this potential, including a greatly reduced number of the Goldstone diagrams [33], a clean separation of the core and valence states, and one set of single-particle states, leading to important simplifications in the calculation of excitation energies and transition matrix elements. Thus, when considering the total energy of different valence states of a one-electron atom, that energy can be written as

$$E = E_v + E_{\mathrm{core}}, \tag{4}$$

where E_{core} is the same for all valence states v. Because the first-order correlation correction to valence removal energies vanishes for a V^{N-1} DF potential, the first nonvanishing corrections are found in second order. The expression for the second-order energy $E^{(2)}$ is [34]

$$E_v^{(2)} = \sum_{mn}\sum_{a} \frac{g_{avmn}(g_{mnav} - g_{mnva})}{\varepsilon_a + \varepsilon_v - \varepsilon_n - \varepsilon_m} + \sum_{n}\sum_{ab} \frac{g_{nvba}(g_{abnv} - g_{abvn})}{\varepsilon_a + \varepsilon_b - \varepsilon_n - \varepsilon_v}. \qquad (5)$$

We use indices a and b to label core states and m and n to designate any excited states. The second-order Coulomb-Breit contribution $B_v^{(2)}$ is obtained from the $E_v^{(2)}$ expression given by Eq. (5) by changing $g_{ijkl} \rightarrow g_{ijkl} + b_{ijkl}$ and keeping only terms that are linear in b_{ijkl}. The b_{ijkl} is a two-particle matrix element of the Breit interaction [35]:

$$B = -\frac{\alpha}{r_{12}}\left[\alpha_1 \cdot \alpha_2 - \frac{1}{2}[\alpha_1 \cdot \alpha_2 - (\alpha_1 \cdot \hat{\mathbf{r}}_{12})(\alpha_2 \cdot \hat{\mathbf{r}}_{12})]\right], \qquad (6)$$

where α_1 is a Dirac matrix, $\hat{\mathbf{r}}_{12} = \mathbf{r}_{12}/r_{12}$, and α is a fine structure constant. The first-order Breit correction is $B_v^{(1)} = \Sigma_a [b_{vava} - b_{vaav}]$.

Even with the elimination of a large set of Goldstone diagrams made possible by the use of the V^{N-1} DF potential, there are still a large number of terms, 52, in the expression for the third-order valence removal energy $E_v^{(3)}$. The corresponding formula for $E_v^{(3)}$ was presented by Blundell *et al.* [9], where the 52 terms were grouped into 12 terms with distinct energy denominators:

$$E_v^{(3)} = E_A^{(3)} + \cdots + E_L^{(L)}. \qquad (7)$$

Expression (7) includes terms with one-, two-, three-, and four-particle sums over virtual states and sums over core states.

The all-order single-double method was discussed previously in Refs. [2–7]. Briefly, we represent the wave function Ψ_v of a one-valence-electron atom as $\Psi_v \cong \Psi_v^{\mathrm{SD}}$ with

$$\Psi_v^{\mathrm{SD}} = \left[1 + \sum_{ma} \rho_{ma} a_m^\dagger a_a + \frac{1}{2}\sum_{mnab} \rho_{mnab} a_m^\dagger a_n^\dagger a_b a_a + \sum_{m \neq v} \rho_{mv} a_m^\dagger a_v + \sum_{mna} \rho_{mnva} a_m^\dagger a_n^\dagger a_a a_v\right]\Phi_v, \qquad (8)$$

where Φ_v is the lowest-order atomic wave function, which is taken to be the frozen-core DF wave function of a state v. We note that we again start from a V^{N-1} DF potential. Substituting the wave function Ψ_v^{SD} into the many-body Schrödinger equation, with the Hamiltonian given by Eqs. (1)–(3), one obtains the coupled equations for the single- and double-excitation coefficients ρ_{mv}, ρ_{ma}, ρ_{mnva}, and ρ_{mnab}. The coupled equations for the excitation coefficients are solved iteratively. We use the resulting excitation coefficients to evaluate hyperfine constants and multipole matrix elements.

The valence E_v^{SD} energy given by

$$E_v^{\mathrm{SD}} = \sum_{ma} \tilde{g}_{vavm}\rho_{ma} + \sum_{mab} g_{abvm}\tilde{\rho}_{mvab} + \sum_{mna} g_{vamn}\tilde{\rho}_{mnva} \qquad (9)$$

does not include a certain part of the third-order contribution. This part of the third-order contribution, $E_{\mathrm{extra}}^{(3)}$, is given in Ref. [5] and needs to be calculated separately. We use our third-order energy code to separate out $E_{\mathrm{extra}}^{(3)}$ and add it to the E_v^{SD}. We drop the index v in the $E_v^{(2)}$, $E_v^{(2)}$, and E_v^{SD} designations in the text and tables below.

We use B-splines [36] to generate a complete set of basis DF wave functions for use in the evaluation of MBPT and all-order expressions. For Tl I and Pb II, we use 40 splines of order $k=7$ for each angular momentum. The basis orbitals are constrained to cavities of radii $R=85$ a.u. and $R=65$ a.u. for Tl I and Pb II, respectively. The cavity radius is chosen large enough to accommodate all $6l_j$ and $5f_j$ orbitals considered here and small enough that 40 splines can approximate inner-shell DF wave functions with good precision.

Results of our energies for the lowest states of neutral Tl and Tl-like Pb ions are summarized in Table I (a complete set of states is given in Tables I and II of the accompanying EPAPS document [37]). The first six rows of Table I give the lowest-order DF energies $E^{(0)}$, second- and third-order Coulomb correlation energies $E^{(2)}$ and $E^{(3)}$, first-order Breit contribution $B^{(1)}$, second-order Coulomb-Breit $B^{(2)}$ corrections, and Lamb shift contribution E_{LS}. We take the sum of these six contributions to be our final third-order RMBPT results, $E_{\mathrm{tot}}^{(3)}$ listed in the seventh row of Table I. We list the all-order SD energies in the row labeled E^{SD} and the part of the third-order energies omitted in the SD calculation in row $E_{\mathrm{extra}}^{(3)}$. We note that E^{SD} contains an $E^{(2)}$ contribution. We take the sum of the six terms $E^{(0)}$, E^{SD}, $E_{\mathrm{extra}}^{(3)}$, $B^{(1)}$, $B^{(2)}$, and E_{LS} to be our final all-order results $E_{\mathrm{tot}}^{\mathrm{SD}}$ listed in the ninth row of Table I. The recommended values from the National Institute of Standards and Technology (NIST) database [38] are given in row labeled E_{NIST}. The differences between our calculations and NIST data, $\delta E^{(3)} = E_{\mathrm{tot}}^{(3)} - E_{\mathrm{NIST}}$ and $\delta E^{\mathrm{SD}} = E_{\mathrm{tot}}^{\mathrm{SD}} - E_{\mathrm{NIST}}$, are given in the last two rows of Table I, respectively.

As expected, the largest correlation contribution to the valence energy comes from the second-order term, $E^{(2)}$. As we shown above, this term is simple to calculate in comparison with $E^{(3)}$ and E^{SD} terms. Thus, we calculate the $E^{(2)}$ term with better accuracy than $E^{(3)}$ and E^{SD} terms.

To increase the accuracy of our calculations we use 50 splines of order $k=9$ for each angular momentum to calculate $E^{(2)}$. The second-order energy $E^{(2)}$ includes partial waves up to $l_{\max}=10$ and is extrapolated to account for contributions from higher partial waves (see, for example, Refs. [39,40]). As an example of the convergence of $E^{(2)}$ with the number of partial waves l, consider the $6p_{1/2}$ state in neutral Tl. Calculations of $E^{(2)}$ with $l_{\max}=8$, 9, and 10 yield $E^{(2)}(6p_{1/2}) = -7682$, -7708, and -7724 cm^{-1}, respectively. Extrapolation of these calculations yields -7737, -7747, and -7753 cm^{-1}, respectively. Therefore, we estimate the numerical uncertainty of $E^{(2)}(6p_{1/2})$ to be approximately 10 cm^{-1}. Similar convergence patterns are found for all other states considered.

Owing to the numerical complexity of the E^{SD} calculation we use $l_{\max}=6$ in our all-order calculations. As we noted above, the second-order $E^{(2)}$ is contained in the E^{SD} value. Therefore, we use our high-precision calculation of $E^{(2)}$ described above to account for the contributions of the higher partial waves. We simply replace $E^{(2)}[l_{\max}=6]$ value with the final high-precision second-order value $E_{\mathrm{final}}^{(2)}$:

TABLE I. Zeroth-order (DF), second-, and third-order Coulomb correlation energies $E^{(n)}$, single-double Coulomb E^{SD}, $E^{(3)}_{\mathrm{extra}}$, and first-order Breit and second-order Coulomb-Breit corrections $B^{(n)}$ to the energies of Tl I and Pb II. The total energies ($E^{(3)}_{\mathrm{tot}}=E^{(0)}+E^{(2)}+E^{(3)}+B^{(1)}+B^{(2)}+E_{\mathrm{LS}}, E^{\mathrm{SD}}_{\mathrm{tot}}=E^{(0)}+E^{\mathrm{SD}}+E^{(3)}_{\mathrm{extra}}+B^{(1)}+B^{(2)}+E_{\mathrm{LS}}$) for Tl I and Pb II are compared with experimental energies E_{NIST} [38], $\delta E=E_{\mathrm{tot}}-E_{\mathrm{NIST}}$. Units: cm^{-1}.

Contr.	$6p_{1/2}$	$6p_{3/2}$	$7s_{1/2}$	$6d_{3/2}$	$5f_{5/2}$	$6p_{1/2}$	$6p_{3/2}$	$7s_{1/2}$	$6d_{3/2}$	$5f_{5/2}$
			Tl I					Pb II		
$E^{(0)}$	−43824	−36636	−21109	−12218	−6863	−114546	−100787	−58728	−46790	−27730
$E^{(2)}$	−7753	−6747	−2024	−958	−98	−9473	−8807	−3911	−4438	−1099
$E^{(3)}$	2534	2414	663	296	25	2947	3007	1331	1354	340
$B^{(1)}$	259	135	26	3	0	460	264	71	35	0
$B^{(2)}$	−421	−291	−46	−8	0	−623	−472	−108	−74	−4
E_{LS}	−2	0	3	0	0	−3	1	10	0	0
$E^{(3)}_{\mathrm{tot}}$	−49205	−41125	−22486	−12886	−6936	−121238	−106794	−61335	−49914	−28493
E^{SD}	−5972	−5446	−1958	−1104	−97	−7239	−6956	−3766	−5129	−1078
$E^{(3)}_{\mathrm{extra}}$	694	807	266	152	16	732	934	510	664	170
$E^{\mathrm{SD}}_{\mathrm{tot}}$	−49266	−41432	−22818	−13175	−6945	−121218	−107016	−62010	−51294	−28642
E_{NIST}	−49264	−41471	−22787	−13146	−6948	−121243	−107162	−61795	−51503	−28729
$\delta E^{(3)}$	59	346	301	260	12	5	368	460	1589	236
δE^{SD}	−2	39	−31	−29	3	25	146	−215	209	87

$$E^{\mathrm{SD}}_{\mathrm{final}}=E^{\mathrm{SD}}+E^{(2)}_{\mathrm{final}}-E^{(2)}[l_{\mathrm{max}}=6].$$

We illustrate this procedure for the $6p_{1/2}$ state. The value of $E^{(2)}(6p_{1/2})$ calculated with 40 splines of order $k=7$ and $l_{\mathrm{max}}=7$ is equal to -7548 cm^{-1}. The value of $E^{(2)}(6p_{1/2})$ calculated with 50 splines of order $k=9$, $l_{\mathrm{max}}=10$, and including extrapolation is equal to -7753 cm^{-1}. The difference between the two values is equal to -205 cm^{-1}. We add this additional contribution to the value of E^{SD} calculated with 40 splines of order $k=7$ and $l_{\mathrm{max}}=6$. A similar value of the additional term (-145 cm^{-1}) is found for the $6p_{3/2}$ state. However, this contribution is much smaller (1–20 cm^{-1}) for all other valence states considered in this work. We give the final results for the all-order energy E^{SD} in Table I.

A lower number of partial waves, $l_{\mathrm{max}}=4$ and 5, is used in a third-order calculation. Since the asymptotic l dependences of the second- and third-order energies are similar (both fall off as l^{-4}), we use the second-order remainder as a guide to estimate the numerical error in the third-order contribution. Using the breakdown of the results for the $E^{(2)}(6p_{1/2})$, with contribution from $l_{\mathrm{max}}>4$ up to ∞ (equal to 9.5%), we estimate the numerical errors for $E^{(3)}(6p_{1/2})$ to be equal to 240 cm^{-1}. A similar value (-180 cm^{-1}) is found for the $6p_{3/2}$ state. Numerical uncertainties are much smaller (1–20 cm^{-1}) for all other states considered in this work. It should be noted that a limited number of partial waves were used by Kozlov *et al.* [15] to calculate the MBPT diagram contributions ($l=5$) and for solving the random-phase approximation equations ($l=4$).

The contribution $E^{(3)}_{\mathrm{extra}}$ given in Table I accounts for the part of the third-order MBPT correction that is not included in the SD energy. The values of $E^{(3)}_{\mathrm{extra}}$ are smaller than the values of $E^{(3)}$ by a factor of 3. As a result, the estimated errors of the sums from $l_{\mathrm{max}}>4$ should be proportionally smaller than the estimated error of the $E^{(3)}$ calculation.

The first-order Breit energies (column $B^{(1)}$ of Table I) include retardation, whereas the second-order Coulomb-Breit energies (column $B^{(2)}$ of Table I) are evaluated using the unretarded Breit operator. The Lamb shift E_{LS} is approximated as the sum of the one-electron self-energy and the first-order vacuum-polarization energy. The vacuum-polarization contribution is calculated from the Uehling potential using the results of Fullerton and Rinker [41]. The self-energy contribution is estimated for the s, $p_{1/2}$, and $p_{3/2}$ orbitals by interpolating among the values obtained by [42–44] using Coulomb wave functions. For this purpose, an effective nuclear charge Z_{eff} is obtained by finding the value of Z_{eff} required to give a Coulomb orbital with the same average $\langle r\rangle$ as the DF orbital. It should be noted that the values of E_{LS} are very small ($E_{\mathrm{LS}}\leqslant 3$ cm^{-1} for Tl I and $E_{\mathrm{LS}}\leqslant 10$ cm^{-1} for Pb II).

The total $E^{(3)}_{\mathrm{tot}}$ in Table I is the sum of six terms, $E^{(0)}$, $E^{(2)}$, $E^{(3)}$, $B^{(1)}$, $B^{(2)}$, and E_{LS}. We find that the correlation corrections to energies in neutral Tl and Tl-like Pb are large, especially for the $6p$ states. For example, $E^{(2)}$ is about 20% of $E^{(0)}$ and $E^{(3)}$ is about 40% of $E^{(2)}$ for the $6p_j$ states of neutral Tl. Despite the evident slow convergence of the perturbation theory expansion, the $6p_{1/2}$ energy from the third-order MBPT calculation is within 0.2% of the measured ionization energy for the $6p_{1/2}$ state of neutral Tl and improves for higher valence states and for Pb^{+} (0.03%). The order of levels changes from Tl to Pb^{+}. For example, the $5f_{5/2}$ and $6f_{5/2}$ states, which are in the 13th and 19th places for neutral Tl, are in the 9th and 15th places for Pb^{+}. In the column $\delta E^{(3)}$ of Table I, we compare our results for the energy levels of interest in Tl I and Pb II with NIST data [38]. The differences $\delta E^{(3)}$ between $E^{(3)}_{\mathrm{tot}}$ and E_{NIST} are generally small, except for

TABLE II. Reduced electric-dipole matrix elements in first, second, third, and all orders of perturbation theory in Tl I and Pb II.

Transition		$Z^{(\mathrm{DF})}$	$Z^{(\mathrm{DF+2})}$	$Z^{(\mathrm{DF+2+3})}$	$Z^{(\mathrm{SD})}$	$Z^{(\mathrm{DF})}$	$Z^{(\mathrm{DF+2})}$	$Z^{(\mathrm{DF+2+3})}$	$Z^{(\mathrm{SD})}$
		Tl I				Pb II			
$6p_{1/2}$	$7s_{1/2}$	2.0484	1.9122	1.6613	1.8200	1.3753	1.2966	1.1715	1.0085
$6p_{3/2}$	$7s_{1/2}$	3.9655	3.6551	3.0780	3.3945	2.6208	2.3611	2.2085	2.0529
$6p_{1/2}$	$6d_{3/2}$	2.7215	2.4497	2.3033	2.3739	2.8176	2.3669	2.1956	2.0661
$6p_{3/2}$	$6d_{3/2}$	1.6334	1.4983	1.3628	1.4193	1.5267	1.3053	1.2081	1.2177
$6p_{3/2}$	$6d_{5/2}$	4.8402	4.4464	3.8568	4.1692	4.5018	3.8655	3.6018	3.4345

the results for the $6p_{3/2}$, $7s_{1/2}$, and nd_j states. One cause of these discrepancies is the limitation of the number of partial waves, $l_{\max}$ in the $E^{(3)}$ calculation. The second cause for these discrepancies is the omission of the fourth- and higher-order correlation corrections in the theoretical values. Another possible cause of the discrepancies is the omission of three-particle interactions in our single-particle model space. The importance of the $6s^2 6d + 6s6p^2$ mixing for Tl I was emphasized by Johnson *et al.* [12].

The total $E^{\mathrm{SD}}_{\mathrm{tot}}$ in Table I is the sum of six terms: $E^{(0)}$, E^{SD}, $E^{(3)}_{\mathrm{extra}}$, $B^{(1)}$, $B^{(2)}$, and E_{LS}. The column labeled δE^{SD} in Table I gives differences between our *ab initio* results and the recommended values [38]. The SD results agree better with the recommended values than the third-order MBPT results, except for the ionization potential of Pb II where the differences with experiment are small for both calculations. Better agreement of the all-order values with experiment demonstrates the importance of the higher-order correlation contributions. Those differences between the present theoretical results and the recommended values that were not improved by the SD method are most probably due to the omission of three-particle interactions mentioned previously.

III. ELECTRIC-DIPOLE MATRIX ELEMENTS, OSCILLATOR STRENGTHS, TRANSITION RATES, AND LIFETIMES IN Tl I AND Pb II

The one-body matrix element of the operator Z is given by [2]

$$Z_{wv} = \frac{\langle \Psi_w | Z | \Psi_v \rangle}{\sqrt{\langle \Psi_v | \Psi_v \rangle \langle \Psi_w | \Psi_w \rangle}}, \tag{10}$$

where Ψ_v is the exact wave function for the many-body "no-pair" Hamiltonian H:

$$H|\Psi_v\rangle = E|\Psi_v\rangle. \tag{11}$$

In MBPT, we expand the many-electron Ψ_v in powers of V_I as

$$|\Psi_v\rangle = |\Psi_v^{(0)}\rangle + |\Psi_v^{(1)}\rangle + |\Psi_v^{(2)}\rangle + |\Psi_v^{(3)}\rangle + \cdots. \tag{12}$$

The denominator in Eq. (10) arises from the normalization condition that starts to contribute in the third order [45]. In the lowest order, we find

$$Z^{(1)}_{wv} = \langle \Psi_w^{(0)} | Z | \Psi_v^{(0)} \rangle = z_{wv}, \tag{13}$$

where z_{wv} is the one-electron corresponding matrix element [46]. Since $\Psi_w^{(0)}$ is a DF function, we use a $Z^{(\mathrm{DF})}$ designation instead of $Z^{(1)}$ below.

The second-order Coulomb correction to the transition matrix element in the DF case with the V^{N-1} potential is given by [47]

$$Z^{(2)}_{wv} = \sum_{na} \frac{z_{an}(g_{wnva} - g_{wnav})}{\varepsilon_a + \varepsilon_v - \varepsilon_n - \varepsilon_w} + \sum_{na} \frac{(g_{wavn} - g_{wanv}) z_{na}}{\varepsilon_a + \varepsilon_w - \varepsilon_n - \varepsilon_v}. \tag{14}$$

The second-order Breit corrections are obtained from Eq. (14) by changing g_{ijkl} to b_{ijkl} [35].

The third-order Coulomb correction is obtained from Eqs. (10) and (12) as

$$Z^{(3)}_{wv} = \langle \Psi_w^{(2)} | Z | \Psi_v^{(0)} \rangle + \langle \Psi_w^{(0)} | Z | \Psi_v^{(2)} \rangle + \langle \Psi_w^{(1)} | Z | \Psi_v^{(1)} \rangle - \frac{1}{2} Z^{(1)}_{wv} [\langle \Psi_v^{(1)} | \Psi_v^{(1)} \rangle + \langle \Psi_w^{(1)} | \Psi_w^{(1)} \rangle], \tag{15}$$

where the last term arises from the normalization condition. The contributions to $Z^{(3)}_{wv}$ were separated as

$$Z^{(3)}_{wv} = Z^{(\mathrm{RPA})} + Z^{(\mathrm{BO})} + Z^{(\mathrm{SR})} + Z^{(\mathrm{norm})} \tag{16}$$

in Ref. [45]. We include the corresponding set of the high-order contributions using the well known random phase approximation (RPA) in $Z^{(\mathrm{RPA})}$ term using the procedure described in Ref. [45]. The subscript BO stands for Brueckner orbitals. The last two terms in Eq. (16) describe structural radiation, $Z^{(\mathrm{SR})}$, and normalization, $Z^{(\mathrm{norm})}$, terms.

In all-order SD calculation, we substitute the all-order SD Ψ_v^{SD} wave function into the matrix element expression given by Eq. (10). In the result, Z^{SD}_{wv} is presented in the following form [2]:

$$Z^{(\mathrm{SD})}_{wv} = \frac{z_{wv} + Z^{(a)} + \cdots + Z^{(t)}}{\sqrt{(1 + N_w)(1 + N_v)}}, \tag{17}$$

where Z_{wv} is the lowest-order (DF) matrix element given by Eq. (13) and the terms $Z^{(k)}$, $k = a \cdots t$ are linear or quadratic functions of the excitation coefficients introduced in Eq. (8). The normalization terms N_w are quadratic functions of the excitation coefficients. As a result of such a procedure, certain sets of many-body perturbation theory terms are summed to all orders.

TABLE III. Reduced electric-dipole matrix elements in Tl I. SD values $Z^{(\mathrm{SD})}$ are compared with theoretical $Z^{(\mathrm{theor})}$ and experimental $Z^{(\mathrm{expt})}$ data given in Ref. [15] and references therein.

Transition	$Z^{(\mathrm{SD})}$	$Z^{(\mathrm{theor})}$	$Z^{(\mathrm{expt})}$
$6p_{1/2}$-$7s_{1/2}$	1.82	1.77	1.81±0.02
$6p_{1/2}$-$6d_{3/2}$	2.37	2.30	2.30±0.09
$6p_{3/2}$-$7s_{1/2}$	3.39	3.35	3.28±0.04
$6p_{3/2}$-$6d_{3/2}$	1.42	1.40	1.38±0.07
$6p_{3/2}$-$6d_{5/2}$	4.17	4.08	4.0±0.2
$7p_{1/2}$-$7s_{1/2}$	5.90	5.96	5.87±0.08
$7p_{1/2}$-$6d_{3/2}$	10.58	10.86	
$7p_{3/2}$-$7s_{1/2}$	7.87	7.98	7.88±0.11
$7p_{3/2}$-$6d_{3/2}$	4.75	4.90	
$7p_{3/2}$-$6d_{5/2}$	14.40	14.88	

The calculation of the transition matrix elements provides another test of the quality of atomic-structure calculations and another measure of the size of correlation corrections. Reduced electric-dipole matrix elements between low-lying states of Tl I and Pb II calculated in the third-order RMBPT and in the all-order SD approximation are presented in Table II. We include only a limited number of transitions in this table to illustrate our results. A complete set of transitions is available as supplementary data in Table III of Ref. [37].

Our calculations of reduced matrix elements in the lowest-, second-, and third-order $Z^{(n)}$ in neutral Tl and Tl-like Pb ion are carried out following the method described above. The lowest order DF value is obtained from Eq. (13). The values of $Z^{(\mathrm{DF+2})}$ are the sum of the second-order correlation correction $Z^{(2)}$ given by Eq. (14) and the DF matrix elements $Z^{(\mathrm{DF})}$. The third-order matrix elements $Z^{(\mathrm{DF+2+3})}$ include the DF values, the second-order $Z^{(2)}$ results, and the third-order $Z^{(3)}$ correlation correction given by the Eq. (16). The term $Z^{(\mathrm{RPA})}$ contains contributions not only from the third-order but also from higher-order RPA corrections as noted above. The BO corrections give the largest contributions to $Z^{(3)}$. The ratio of $Z^{(\mathrm{BO})}$ and $Z^{(\mathrm{DF})}$ terms is about 5%–15%. The term $Z^{(\mathrm{RPA})}$ is about 5%–10% of the $Z^{(\mathrm{DF})}$ term. The smallest contributions (about 1%) are from the structural radiation, $Z^{(\mathrm{SR})}$, and normalization, $Z^{(\mathrm{norm})}$, terms. The basis set used here is the same as in the calculation of the energy contributions. We find correlation corrections $Z^{(2+3)}$ to be very large, 10%–30% for many cases. All results given in Table II are obtained using length form of the matrix elements. Length-form and velocity-form matrix elements differ typically by 1%–10% for the DF matrix elements and 1%–3% for the second-order matrix elements.

The electric-dipole matrix elements evaluated in the all-order SD approximation are given in the columns labeled $Z^{(\mathrm{SD})}$ [Eq. (17)] of Table II. The SD matrix elements $Z^{(\mathrm{SD})}$ include $Z^{(3)}$ completely, along with important fourth- and higher-order corrections. The fourth-order corrections omitted from the SD matrix elements were discussed recently by [48]. The $Z^{(\mathrm{SD})}$ values are smaller than the $Z^{(\mathrm{DF+2})}$ values and larger than the $Z^{(\mathrm{DF+2+3})}$ values for all Tl I transitions given in Table II and some of the Pb II transitions.

In Table III, we compare our SD data $Z^{(\mathrm{SD})}$ for reduced electric-dipole matrix elements in Tl I with theoretical $Z^{(\mathrm{theor})}$ and experimental $Z^{(\mathrm{expt})}$ data given by Kozlov *et al.* [15] and references therein. Our SD data are in excellent agreement with experimental and theoretical data from Refs. [14,15] obtained by combining MBPT and CI methods.

Transition rates A_r (s^{-1}), oscillator strengths (f), and line strengths S (a.u.) for the 72 transitions in Tl I and Pb II are too voluminous to include here. They are tabulated and compared with experiment in Tables IV–VI of the accompanying EPAPS document [37], respectively. We use theoretical energies obtained in the SD approximation $E_{\mathrm{tot}}^{\mathrm{SD}}$ and the SD matrix elements to calculate those values using well-known expressions (see, for example, [38]). For convenience, we present wavelengths for all transitions in Tables IV–VI of [37]. The largest oscillator strengths agree with experimental results within the corresponding uncertainties in many cases. Our SD results for the wavelengths are in excellent agreement with experimental wavelengths; the discrepancies are about 0.01%–0.1%.

We calculate lifetimes of the np_j, $ns_{1/2}$ ($n=7-9$), nd_j ($n=6-8$), and $nf_{5/2}$ ($n=5-6$) states in Tl I and Pb II using SD results for dipole matrix elements and energies. We compare these lifetimes $\tau^{(\mathrm{SD})}$ with available experimental measurements in Table IV. The experimental data for Tl I are from Refs. [24,25] and the Pb II data are from Ref. [21] and references therein. Experimental results for τ^{expt} were given in Refs. [25,21] with 5% and 10% errors, respectively. Our present values are in excellent agreement with experimental results except for one case, the $7s_{1/2}$ state in Pb II. This is

TABLE IV. Lifetimes τ of the nl levels in Tl I and Pb II in ns. The SD data are compared with experimental results for Tl I from Refs. [24,25] and for Pb II from Ref. [21] and references therein.

Level	$\tau^{(\mathrm{SD})}$	τ^{expt}	Level	$\tau^{(\mathrm{SD})}$	τ^{expt}
	Tl I, $Z=81$			Pb II, $Z=82$	
$7s_{1/2}$	7.43	7.45±0.2	$7s_{1/2}$	1.68	7.2±0.9
$7p_{1/2}$	61.8	63.1±1.7	$7p_{1/2}$	16.9	15.2±1.7
$7p_{3/2}$	47.3	48.6±1.3	$7p_{3/2}$	10.4	10.3±1.2
$8p_{1/2}$	177.6	184.1±4.4	$7d_{3/2}$	4.05	3.4±0.4
$8p_{3/2}$	123.5	127.7±4.9	$5f_{5/2}$	4.43	5.9±0.6
$9p_{1/2}$	375.1	391.1±21.8	$6f_{5/2}$	11.5	11.6±1.5
$9p_{3/2}$	251.3	273.6±13.5			

TABLE V. Reduced matrix elements of the electric-quadrupole and magnetic-dipole operators in first, second, third, and all orders of perturbation theory in Tl I and Pb II.

Transition		$Z^{(\mathrm{DF})}$	$Z^{(\mathrm{DF+2})}$	$Z^{(\mathrm{DF+2+3})}$	$Z^{(\mathrm{SD})}$	$Z^{(\mathrm{DF})}$	$Z^{(\mathrm{DF+2})}$	$Z^{(\mathrm{DF+2+3})}$	$Z^{(\mathrm{SD})}$
		Tl I				Pb II			
				Electric-quadrupole transitions					
$6p_{1/2}$	$6p_{3/2}$	15.2950	15.4716	12.4935	13.1672	9.1374	9.1936	8.1941	8.2784
$6p_{1/2}$	$7p_{3/2}$	7.1787	7.2776	5.7125	6.3019	4.0244	4.1182	3.8682	4.0294
$6p_{3/2}$	$7p_{1/2}$	15.8767	15.9107	11.7256	13.2822	8.0165	8.0706	7.3327	7.6247
$7p_{1/2}$	$7p_{3/2}$	127.7551	128.1446	110.4106	114.0819	53.1141	53.1487	47.8697	48.6564
				Magnetic-dipole transitions					
$6p_{1/2}$	$6p_{3/2}$	1.1354	1.1358	1.0959	1.1366	1.1372	1.1375	1.0963	1.1371
$6p_{1/2}$	$7p_{3/2}$	0.1022	0.1016	0.0771	0.1020	0.0971	0.0965	0.0832	0.0971
$6p_{3/2}$	$7p_{1/2}$	0.1228	0.1218	0.1137	0.1077	0.1169	0.1160	0.1197	0.1161
$7p_{1/2}$	$7p_{3/2}$	1.1387	1.1380	1.1345	1.1384	1.1391	1.1393	1.1261	1.1379

unexpected since we have perfect agreement between τ^{SD} and τ^{expt} from Ref. [24] for the $7s_{1/2}$ state in Tl I. As one can see from Table IV, the lifetime values in Tl I are larger than the lifetime values in Pb II by a factor of 4. In this case, it seems strange that τ^{expt} for the $7s_{1/2}$ state in Pb II is almost identical with τ^{expt} for the $7s_{1/2}$ state in Tl I.

IV. ELECTRIC-QUADRUPOLE AND MAGNETIC-DIPOLE TRANSITIONS IN Tl I AND Pb II

Reduced matrix elements of the electric-quadrupole $(E2)$ and magnetic-dipole $(M1)$ operators in lowest, second, third, and all orders of perturbation theory are given in Table V for Tl I and Pb II. Detailed descriptions of the calculations of the reduced matrix elements of the $E2$ and $M1$ operators in lowest and second orders of perturbation theory were given by Safronova *et al.* [49]. Third-order and all-order calculations are done in the same way as the calculations of $E1$ matrix elements. In Table V, we present $E2$ and $M1$ matrix elements in the $Z^{(\mathrm{DF})}$, $Z^{(\mathrm{DF+2})}$, $Z^{(\mathrm{DF+2+3})}$, and $Z^{(\mathrm{SD})}$ approximations for $6p_{1/2}$-$6p_{3/2}$, $6p_{1/2}$-$7p_{3/2}$, $7p_{1/2}$-$6p_{3/2}$, and $7p_{1/2}$-$7p_{3/2}$ transitions in Tl I and Pb II. These four transitions in Tl I were investigated for the first time in [29]. The importance of the Breit contribution to the calculation of the np_j-$n'p_{j'}$ matrix elements in Tl I was underlined in Ref. [29]. We also found that the second-order Breit contributions are larger than the Coulomb contributions, which are unusually small for the above-mentioned $E2$ and $M1$ transitions. As a result, the difference between $Z^{(\mathrm{DF})}$ and $Z^{(\mathrm{DF+2})}$ presented in Table V is rather small, about 0.3%–1%. The largest Coulomb contribu-

TABLE VI. Wavelengths λ (Å) and transition rates for electric-quadrupole A_r^{E2} and magnetic-dipole A_r^{M1} (s^{-1}) transitions in Tl I and Pb II calculated in the SD approximation. The SD data (a) are compared with theoretical calculations given in Refs. [29] (b) and [28] (c). Numbers in brackets represent powers of 10.

Transition			λ	A_r^{E2}	A_r^{M1}
			Tl I		
$6p_{1/2}$	$6p_{3/2}$	*a*	12862	0.1379	4.094
		b		0.158	4.085
		c		0.1978	4.268
$6p_{1/2}$	$7p_{3/2}$	*a*	2861	58.05	2.996
		b		55.2	3.31
$6p_{3/2}$	$7p_{1/2}$	*a*	3819	121.5	2.810
		b		72.8	2.18
$7p_{1/2}$	$7p_{3/2}$	*a*	99900	3.66[−4]	8.765[−3]
		b		3.69[−4]	8.706[−3]
			Pb II		
$6p_{1/2}$	$6p_{3/2}$	*a*	7074	1.083	24.63
		c		1.365	25.2
$6p_{1/2}$	$7p_{3/2}$	*a*	1298	1236	29.13
$6p_{3/2}$	$7p_{1/2}$	*a*	1663	2557	395.3
$7p_{1/2}$	$7p_{3/2}$	*a*	35549	0.0117	0.1944

tions arise from the Bruekner-orbital $Z^{(\mathrm{BO})}$ correction which is especially large for $E2$ matrix elements in Tl; the ratio $Z^{(\mathrm{BO})}/Z^{(\mathrm{DF})}$ is about 0.13–0.25.

Transition rates for the $E2$ transitions A_r^{E2} and $M1$ transitions A_r^{M1} (s^{-1}) in Tl I and Pb II calculated in the SD approximation are presented in Table VI. The SD data (a) are compared with theoretical calculations given in Refs. [28,29] (b, c). The differences between our results and those from [28,29] for A_r^{E2} and A_r^{M1} can be explained by the additional correlation corrections taken into account in our calculations.

V. HYPERFINE CONSTANTS FOR NEUTRAL THALLIUM

Calculations of hyperfine constants follow the same pattern as calculations of the reduced $E1$, $E2$, and $M1$ matrix elements described in the previous section. The magnetic moment and nuclear spin used in the present calculations are taken from [50].

Contributions to the SD values for the $6p_j$, $6d_j$, and $7s_{1/2}$ states in ^{205}Tl are given in Table VII of the accompanying EPAPS document [37]. In Table VII, we list hyperfine constants A for Tl and compare our values with available theoretical and experimental data from Refs. [15,29,30] and references therein. In this table, we present the first-order $A^{(\mathrm{DF})}$ and all-order $A^{(\mathrm{SD})}$ values for the np_j levels with n=6–9, $ns_{1/2}$ levels with n=7–9, and $6d_j$ levels. The largest disagreements between our SD data and the experimental values occur for $6p_{3/2}$ and $8d_{5/2}$ states. The correlation correction for $6p_{3/2}$ state is of the same order of magnitude as the DF value and has an opposite sign. With such a cancellation it is difficult to calculate $A(6p_{3/2})$ accurately. The best agreement with experimental measurements is found for the $6p_{1/2}$ and $7p_{1/2}$ states; the differences of our A values with experiment for these states are 0.4% and 0.3%, respectively. For other states ($7s_{1/2}$, $7p_{3/2}$, $6d_j$, $8p_j$, and $9p_j$) the discrepancies range from 2% to 7%. We note that the correlation corrections to A values are very large for essentially all of the states. Yet we find that the SD method produces results in good agreement with experiment in most cases.

TABLE VII. Hyperfine constants A (in MHz) for the np_j with n=6–9, $ns_{1/2}$ with n=7–9, and $6d_j$ levels in ^{205}Tl ($I=1/2, \mu=1.638\,213\,5$). The SD data are compared with theoretical and experimental results from Ref. [15] (a), Ref. [29] (b), Ref. [30] (c), and Ref. [31] (d).

Level	$A^{(\mathrm{DF})}$	$A^{(\mathrm{SD})}$	$A^{(\mathrm{theor})}$	$A^{(\mathrm{expt})}$
$6p_{1/2}$	17414	21390	21663[a]	21310.8±0.0[c]
$6p_{3/2}$	1302	353	248[a]	265.0±0.0[c]
$7s_{1/2}$	7381	12596	12666[a]	12297.2±1.6[d]
$7p_{1/2}$	1942	2248	2193[a]	2155.5±0.6[c]
$7p_{3/2}$	187.9	294.3	295[a]	311.4±0.3[c]
$6d_{3/2}$	20.7	−42.1	−41[a]	−42.9±0.4[d]
$6d_{5/2}$	8.66	215.4	183[a]	226.9±0.3[d]
$8s_{1/2}$	2479	3908	4320[b]	3870.8±1.4[d]
$8p_{1/2}$	730	836	706[c]	788.5±0.9[c]
$8p_{3/2}$	72.1	122	61[c]	130.2±0.5[c]
$9s_{1/2}$	1127	1657	1900[b]	1779.4±1.2[d]
$9p_{1/2}$	356	401		378.4±0.8[c]
$9p_{3/2}$	35.7	62.1		67.1±0.2[c]
$7d_{3/2}$	11.0	−45.6		−56.3±0.6[d]
$7d_{5/2}$	4.55	135		180.2±0.2[d]
$8d_{3/2}$	6.10	−32.0		−42.3±0.4[d]
$8d_{5/2}$	2.53	83.1		130.6±0.2[d]

[a] Reference [15].
[b] Reference [29].
[c] Reference [30].
[d] Reference [31].

VI. CONCLUSION

In summary, a systematic relativistic MBPT study of the energies of $6s^2np_j$ (n=6–9), $6s^2ns_{1/2}$ (n=7–9), $6s^2nd_j$ (n=6–8), and $6s^2nf_{5/2}$ (n=5–6) states in Tl I and Pb II is presented. The energy values are in good agreement with existing experimental energy data and provide a theoretical reference database for the line identification. A systematic all-order SD study of the reduced matrix elements, oscillator strengths, and transition rates for the 72 possible $6s^2nl_j$-$6s^2n'l'_{j'}$ electric-dipole transitions is conducted. Electric-quadrupole and magnetic-dipole matrix elements are evaluated to calculate the $6s^2np_{1/2}$-$6s^2np_{3/2}$ transition rates. Hyperfine constants are presented for $6s^2np_j$ (n=6–9), $6s^2ns_{1/2}$ (n=7–8), and $6s^2nd_j$ (n=6–8) states in ^{205}Tl isotopes. We believe that our energies and transition rates will be useful in analyzing existing experimental data and in planning future measurements.

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