Relativistic many-body calculations of the Stark-induced amplitude of the $6P_{1/2} - 7P_{1/2}$ transition in thallium

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Stark-induced amplitudes for the $6P_{1/2} - 7P_{1/2}$ transition in Tl I are calculated using the relativistic SD approximation in which single and double excitations of Dirac-Hartree-Fock levels are summed to all orders in perturbation theory. Our SD values $\alpha_S = 368\ a_0^3$ and $|\beta_S| = 298\ a_0^3$ are in good agreement with the measurements α_S=377(8) a_0^3 and $\beta_S = 313(8)\ a_0^3$ by D. DeMille, D. Budker, and E. D. Commins [Phys. Rev. A**50**, 4657 (1994)]. Calculations of the Stark shifts in the $6P_{1/2} - 7P_{1/2}$ and $6P_{1/2} - 7S_{1/2}$ transitions are also carried out. The Stark shifts predicted by our calculations agree with the most accurate measured values within the experimental uncertainties for both transitions.

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I. INTRODUCTION

The 293 nm $6P_{1/2} - 7P_{1/2}$ transition in thallium has been studied extensively both experimentally and theoretically because of its connection to atomic parity nonconservation (PNC). Although $6P_{1/2} - 7P_{1/2}$ is nominally a magnetic dipole (M1) transition, there is also an electric dipole (E1) component arising from the weak interaction mediated by Z_0 exchange between the nucleus and bound electrons. The weak E1 component of the $6P_{1/2} - 7P_{1/2}$ transition is calibrated using the Stark-induced amplitude β_S. Measured values of both $\alpha_S = 2.01(4)\times10^{-5}\mu_B$ cm/V and $\beta_S = 1.67(4)\times10^{-5}\mu_B$ cm/V for the $6P_{1/2} - 7P_{1/2}$ transition in thallium were reported by DeMille et al. [1] and found to be in substantial disagreement with earlier measurements $\alpha_S = 1.31(6)\times10^{-5}\mu_B$ cm/V and $\beta_S = 1.09(5)\times10^{-5}\mu_B$ cm/V by Tanner and Commins [2]. It was pointed out in Ref. [3] that both the Stark shift and the Stark-induced amplitude could be measured in the same transition in which PNC is measured [4].

High-precision measurements of the Stark shift within the 378 nm $6P_{1/2} - 7S_{1/2}$ E1 transition in atomic thallium were recently reported by Doret et al. [3]. The result $\Delta\nu_S$ = -103.23(39) kHz/(kV/cm)^2 had higher accuracy by factor of 15 than earlier measurements [1, 5]. The earlier value for the $6P_{1/2} - 7S_{1/2}$ transition from Ref. [1] was $\Delta\nu_S$ = -112(6) kHz/(kV/cm)^2.

In the present paper, we carry out relativistic all-order calculations of Stark-induced amplitudes α_S and β_S for the $6P_{1/2} - 7P_{1/2}$ transition as well as Stark shifts within both $6P_{1/2} - 7S_{1/2}$ and $6P_{1/2} - 7P_{1/2}$ transitions in atomic thallium. The calculations are carried out using the relativistic SD all-order method in which single and double excitations of Dirac-Hartree-Fock (DHF) wave functions are summed to all orders in perturbation theory. Recently, lifetimes, E1, E2, and M1 transition rates, hyperfine constants, and excitation energies of the nP_J, $nS_{1/2}$, and nD_J states in neutral thallium were evaluated by Safronova et al. [6], using both relativistic many-body perturbation theory (MBPT) and the relativistic SD method. The SD calculations were found to be in excellent agreement with the best available experimental data.

II. STARK-INDUCED AMPLITUDE OF THE $6P_{1/2} - 7P_{1/2}$ TRANSITION IN THALLIUM

Stark-induced scalar and vector polarizabilities α_S and β_S for transitions in Na, K, Rb, Cs, and Fr were calculated in the SD approximation by Safronova et al. [7]. Following the procedure used in [6], we treat Tl I as a one-valence-electron atom with a Hg-like core and evaluate the Stark-induced scalar and vector polarizabilities α_S and β_S following the procedures used in [7] for alkali-metal atoms.

The scalar and vector polarizabilities α_S and β_S for the transitions between the ground state $6P_{1/2}$ and the

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TABLE I: The contributions to Stark-induced scalar polarizability α_S for the $6p_{1/2}-7p_{1/2}$ transition in Tl I. The corresponding energy differences and electric-dipole reduced matrix elements are also listed. All values are given in a.u.

Contribution	nlj	$E_{6P_{1/2}}-E_{nlj}$	$E_{7P_{1/2}}-E_{nlj}$	$Z_{7P_{1/2},nlj}$	$Z_{nlj,6P_{1/2}}$	α_S
$\alpha_S^{\rm main}(nS)$	$7S_{1/2}$	-0.120640	0.035004	6.016	-1.827	-37.2
	$8S_{1/2}$	-0.176539	-0.020895	-6.215	-0.535	-29.7
	$9S_{1/2}$	-0.196680	-0.041036	-1.274	-0.298	-1.9
	$10S_{1/2}$	-0.206387	-0.050743	-0.656	-0.200	-0.5
$\alpha_S^{\rm tail}(nS)$						-2.0
$\alpha_S^{\rm main}(nD_{3/2})$	$6D_{3/2}$	-0.164565	-0.008921	-10.703	-2.334	492.0
	$7D_{3/2}$	-0.191418	-0.035774	4.821	-1.101	-29.4
	$8D_{3/2}$	-0.203543	-0.047899	2.377	-0.672	-6.9
	$9D_{3/2}$	-0.210040	-0.054396	1.535	-0.476	-2.8
$\alpha_S^{\rm tail}(nD_{3/2})$						-14.1
$\delta\alpha_S^{\rm core}$						0.4
Total						368

excited state $7P_{1/2}$ in Tl I are calculated as

$$\alpha_{\rm S} = \sum_n \left[I_\alpha(nS_{1/2}) - I_\alpha(nD_{3/2})\right], \quad (1)$$

$$\beta_{\rm S} = \sum_n \left[I_\beta(nS_{1/2}) + \frac{1}{2} I_\beta(nD_{3/2})\right], \quad (2)$$

where

$$I_\alpha(nlj) = \frac{1}{6}\left[\frac{Z_{7P_{1/2},nlj}Z_{nlj,6P_{1/2}}}{E_{6P_{1/2}}-E_{nlj}} + \frac{Z_{7P_{1/2},nlj}Z_{nlj,6P_{1/2}}}{E_{7P_{1/2}}-E_{nlj}}\right]$$

and

$$I_\beta(nlj) = \frac{1}{6}\left[\frac{Z_{7P_{1/2},nlj}Z_{nlj,6P_{1/2}}}{E_{7P_{1/2}}-E_{nlj}} - \frac{Z_{7P_{1/2},nlj}Z_{nlj,6P_{1/2}}}{E_{6P_{1/2}}-E_{nlj}}\right].$$

The quantities Z_{wv} in the above equations are electric-dipole reduced matrix elements. The all-order SD matrix elements are calculated as [6]

$$Z_{wv} = \frac{z_{wv} + Z^{(a)} + \cdots + Z^{(t)}}{\sqrt{(1+N_w)(1+N_v)}}, \quad (3)$$

where z_{wv} is the lowest-order (DHF) matrix element and the terms $Z^{(k)}$, $k = a\cdots t$ are linear or quadratic function of the SD excitation coefficients. The normalization terms N_w are quadratic functions of the excitation coefficients. The SD matrix elements include all MBPT corrections through third order together with important classes of forth- and higher-order corrections.

The calculation of the $\alpha_{\rm S}$ is divided into three parts:

$$\alpha_{\rm S} = \alpha_S^{\rm main} + \alpha_S^{\rm tail} + \delta\alpha_S^{\rm core}, \quad (4)$$

where $\alpha_S^{\rm main}$ is the dominant contribution from states near the valence state, $\delta\alpha_S^{\rm core}$ is the contribution from core-excited autoionizing states, and $\alpha_S^{\rm tail}$ is the remainder from highly excited one-electron states. Thus, we write,

$$\alpha_S^{\rm main} = \sum_{n=7}^{10} I_\alpha(nS_{1/2}) - \sum_{n=6}^{9} I_\alpha(nD_{3/2}), \quad (5)$$

$$\delta\alpha_S^{\rm core} = \sum_{n=1}^{6} I_\alpha(nS_{1/2}) - \sum_{n=3}^{5} I_\alpha(nD_{3/2}),$$

$$\alpha_{\rm tail} = \sum_{n=11}^{N} I_\alpha(nS_{1/2}) - \sum_{n=10}^{N} I_\alpha(nD_{3/2}),$$

where N is the number of the finite basis set states. The calculation of β_S is conducted in the same way.

We use B-splines [8] to generate a complete set of DHF basis orbitals for use in the evaluation of all electric-dipole matrix elements. Here, we use here $N = 50$ splines (compared with 40 used in [6]) for each angular momentum. The basis orbitals are constrained to a cavity of radius $R = 90$ (a.u.). The size of the cavity is taken to be large enough to fit all of the states needed for the calculation of the main terms for all of the polarizabilities calculated in this work. Furthermore, we use the Breit-Dirac-Hartree-Fock (BDHF) approximation here instead of the Dirac-Hartree-Fock (DHF) approximation used in Ref. [6] because we found that the Breit contributes about 1% to the values of α_S and β_S at the DHF level. The BDHF approximation includes the the one-body part of the Breit interaction in the DHF equation (for more detail see Refs. [9, 10]). The one-body part of the Breit interaction is also included in the equations that generate DHF basis orbitals. In fact, all of the calculations in this work, including the calculations of the tail contributions, are done with same basis set.

The sums over n in Eqs. (1, 2) converge rapidly; therefore, only main term contributions have to be calculated accurately. We calculate main terms for α_S and β_S using SD matrix elements and experimental energies [11]. The

TABLE II: The contributions to Stark-induced vector polarizability β_S(a.u.) for the $6P_{1/2} - 7P_{1/2}$ transition in Tl I.

Contribution	nlj	β_S
$\beta_S^{\rm main}(nS)$	$7S_{1/2}$	-67.5
	$8S_{1/2}$	-23.4
	$9S_{1/2}$	-1.2
	$10S_{1/2}$	-0.3
$\beta_S^{\rm tail}(nS)$		-0.9
$\beta_S^{\rm main}(nD_{3/2})$	$6D_{3/2}$	-220.7
	$7D_{3/2}$	10.1
	$8D_{3/2}$	2.1
	$9D_{3/2}$	0.8
$\beta_S^{\rm tail}(nD_{3/2})$		3.3
$\delta\beta_S^{\rm core}$		0.0
Total		-298

TABLE III: The SD all-order Stark-induced scalar and vector polarizabilities for the $6P_{1/2} - 7P_{1/2}$ transition in Tl are compared with measurements by Tanner and Commins [2] - *a* and DeMille et al. [1] - *b*.

	α_S	$\lvert\beta_S\rvert$
This work	368	298
Expt.[a]	247±12	198±10
Expt.[b]	377±8	313±8

contribution of the remainder, while small, is not negligible for $n < 20$ and is calculated in the random-phase approximation (RPA) for these states. It is essentially zero for larger n and is evaluated in the DHF approximation for $n \geq 20$ without loss of accuracy. We find that it is essential to use RPA approximation to evaluate the tail contribution for α_S as DHF calculation significantly overestimates the tail yielding $-25\ a_0^3$ while RPA calculation gives $-16\ a_0^3$. DHF approximation is expected to significantly overestimate the value of the tail contribution as it significantly overestimates the value of the main term. The autoionizing contribution is very small, 0.1%, for α_S and is negligible at the present level of accuracy for β_S. It is evaluated in the DHF approximation.

In Tables I and II, we present the details of our α_S and β_S calculations for the transition between the ground state $6P_{1/2}$ and the excited $7P_{1/2}$ state in Tl I. We separate the contributions from the nS and $nD_{3/2}$ terms given by the Eqs. (1, 2). Furthermore, the contribution from each n in the Eq. (5) for the main term is given separately in the corresponding (nlj) row to demonstrate rapid convergence of the sums in Eqs. (1, 2). The tail contributions for nS and $nD_{3/2}$ sums are listed in the rows following the corresponding main term. The total contribution from the core-excited autoionizing states are listed in rows labeled $\delta\alpha_S^{\rm core}$ and $\delta\beta_S^{\rm core}$ of Tables I and II, respectively.

We list the values of the reduced electric-dipole matrix elements and energies used in the calculation of α_S and β_S in Table I. Since the same matrix elements and energies contribute to α_S and β_S, we do not repeat the energy and matrix element values in Table II. We use recommended values of energies from the National Institute of Standards and Technology (NIST) database [11] when calculating all of the polarizability values in this work. The corresponding energy differences are listed in columns two and three of Table I. Electric-dipole matrix elements evaluated using the SD all-order method (Eq. (3)) are given in columns labeled Z_{wv}. It should be noted that the values of Z_{wv} given in Table I generally differ by 0.4–1.0% from the values of Z_{wv} presented in Ref. [6]. These differences arise because we include the Breit interaction here and, to the lesser extent, because of more accurate basis set used in the present work.

We also conducted a semiempirical scaling procedure, described for example, in Refs. [7, 12], for three transitions, $7S - 7P_{1/2}$, $7S - 7P_{3/2}$, and $7P_{1/2} - 6D_{3/2}$. These transitions give significant contributions to the α_S and β_S as well as Stark shifts that we discuss below. The scaling is carried out by multiplying the values of the corresponding single valence excitation coefficients by the ratio of the theoretical and experimental correlation energies and repeating the calculation of the matrix elements with the modified excitation coefficients. Study of the breakdown of the correlation corrections showed that the dominant contribution to the correlation correction to the values of these matrix elements comes from a single term that contains only single valence excitation coefficients. It has been shown that the scaling works effectively in such cases (see, for example [13, 14]) since it is specifically aimed at correcting the dominant contribution. In the case of the $7S - 7P$ transitions, the correlation breakdown is the same as in Cs, where the scaled values are in excellent agreement with the high-precision experiment. The scaling can not be applied to improve the values of the $6D_{3/2} - 6P_{1/2}$ matrix element as the the corresponding correlation correction is not dominant, leading to possibly reduced accuracy of this transition in comparison to $7S - 7P_{1/2}$, $7S - 7P_{3/2}$, and $7P_{1/2} - 6D_{3/2}$ ones.

We find that the contribution of the $6D_{3/2}$ term from Eqs. (1, 2) dominates the values of α_S and β_S. While all valence terms contributing to α_S have the same sign, there is significant cancellation of terms contributing to β_S. In fact, all nS terms contribute with the sign opposite to that of the $nD_{3/2}$ terms with the exception of the first ones, $7S$ and $6D_{3/2}$. Owing to this cancellation, our value of β_S may be somewhat less accurate than the value of α_S.

In Table III, we compare our results for the Stark-induced scalar and vector polarizabilities for the $6P_{1/2} - 7P_{1/2}$ transition in Tl with experimental measurements by DeMille et al. [1] and Tanner and Commins [2]. The conversion factor between the units of (μ_B/c) (cm/V) used in Refs. [1, 2] and atomic units used in the present work is $10^{-2}\alpha E_h/2ea_0$=1.8762×10^7, where E_h is Hartree

energy. Our results support the measurements of DeMille et al. [1] and clearly disagree with the measurements reported by Tanner and Commins [2]. Our value of α_S is nearly within the experimental uncertainty of the Ref. [1] measurement and our value of β_S differs from the central experimental value of Ref. [1] by 2σ. As we discussed above, accurate calculation of β_S is more difficult owing to the cancellation of the nS and $nD_{3/2}$ contributions.

III. STARK SHIFT WITHIN THE $6P_{1/2} - 7S_{1/2}$ AND $6P_{1/2} - 7P_{1/2}$ TRANSITIONS IN ATOMIC THALLIUM

We calculate the Stark shifts within the $6P_{1/2} - 7S_{1/2}$ and $6P_{1/2} - 7P_{1/2}$ transitions as a differences of scalar dipole polarizabilities α of the $6P_{1/2}$ ground state and the $7S_{1/2}$ or $7P_{1/2}$ excited states. The expression for α is given by (see, for example, Refs. [5, 15]):

$$\alpha(n_0P_{1/2}) = \sum_n \left[I_S(nS_{1/2}) + I_S(nD_{3/2})\right]$$
$$\alpha(n_0S_{1/2}) = \sum_n \left[I_S(nP_{1/2}) + I_S(nP_{3/2})\right],$$

where

$$I_S(nlj) = \frac{1}{3}\frac{Z^2_{n_0l_0j_0,nlj}}{E_{nlj} - E_{n_0l_0j_0}}.$$

Our results for the $6P_{1/2}$, $7P_{1/2}$, and $7S_{1/2}$ polarizabilities are given in Tables IV and V where we use the same designations as in the previous section. The polarizability $\alpha_{\rm core}$ of the Hg-like ionic core is also evaluated using RPA approximation. A more detailed discussion for the $\alpha_{\rm core}$ in Na, K, Rb, Cs, and Fr atomic systems is found in Ref. [7]. We note that contributions from $\alpha_{\rm core}$ cancel when we evaluate the Stark shift for a transition.

Only the polarizability contributions are listed in Table IV, since all relevant energies and electric-dipole matrix elements are already listed in Table I. The calculation of the $7S_{1/2}$ polarizabilities involves the calculation of other series of the matrix elements, $7S - nP_{1/2}$ and $7S - nP_{3/2}$. We list those values, calculated using SD all-order method, together with the corresponding energy differences taken from [11] in Table V.

The value of the $6P_{1/2}$ polarizability has two dominant (and nearly equal) contributions, from $6P_{1/2}-6D_{3/2}$ and $6P_{1/2} - 7S$ transitions. The value of the $7P_{1/2}$ polarizability is dominated by the contribution from $7P_{1/2}-6D_{3/2}$ transition. The value of the $7S$ polarizability is dominated entirely by the contributions from both $7S-7P$ transitions. As we have discussed above, we conducted more accurate calculation of the $7P_{1/2} - 6D_{3/2}$, $7S-7P_{1/2}$, and $7S-7P_{3/2}$ electric-dipole matrix elements by rescaling the single valence excitation coefficients with the correct value of the correlation energy leading to more accurate evaluation of the dominant contributions to $7S$ and $7P_{1/2}$ polarizabilities. Our value for the $\alpha(6P_{1/2}) = 50.4$ $a_0{}^3$ is in good agreement with the theoretical result 49.2 $a_0{}^3$ by Kozlov et al. [16].

In Table VI, we compare our results for the Stark shift in the $6P_{1/2} - 7S_{1/2}$ and $6P_{1/2} - 7P_{1/2}$ transitions with

TABLE IV: The contributions to the scalar dipole $6P_{1/2}$ and $7P_{1/2}$ polarizabilities α(a.u.) in Tl.

Contribution	nlj	$\alpha(6P_{1/2})$	$\alpha(7P_{1/2})$
$\alpha^{\rm main}(nS)$	$7S_{1/2}$	9.2	-345
	$8S_{1/2}$	0.5	616
	$9S_{1/2}$	0.2	13
	$10S_{1/2}$	0.1	3
$\alpha^{\rm tail}(nS)$		0.7	6
$\alpha^{\rm main}(nD_{3/2})$	$6D_{3/2}$	11.0	4280
	$7D_{3/2}$	2.1	217
	$8D_{3/2}$	0.7	39
	$9D_{3/2}$	0.4	14
$\alpha^{\rm tail}(nD_{3/2})$		4.7	47
$\alpha^{\rm core}$		24.1	24
$\delta\alpha_S^{\rm core}$		-3.3	0
Total		50.4	4915

TABLE V: The contributions to the scalar dipole $7S$ polarizability α in Tl. The energy differences and absolute values of the electric-dipole reduced matrix elements for relevant transitions are also listed. All values are given in a.u.

Contribution	nlj	$E_{nlj} - E_{7S}$	$\lvert Z_{7S,nlj}\rvert$	$\alpha(7S_{1/2})$
$\alpha^{\rm main}(nP_{1/2})$	$6P_{1/2}$	-0.120640	1.826	-9.2
	$7P_{1/2}$	0.035004	6.016	344.7
	$8P_{1/2}$	0.067847	0.706	2.5
	$9P_{1/2}$	0.081574	0.296	0.4
$\alpha^{\rm tail}(nP_{1/2})$				0.4
$\alpha^{\rm main}(nP_{3/2})$	$6P_{3/2}$	-0.085134	3.397	-45.2
	$7P_{3/2}$	0.039565	8.063	547.7
	$8P_{3/2}$	0.069545	1.474	10.4
	$9P_{3/2}$	0.082401	0.713	2.1
$\alpha^{\rm tail}(nP_{3/2})$				3.0
$\alpha^{\rm core}$				24.1
Total				880.8

TABLE VI: The all-order values of the ground and excited state polarizability differences are compared with experimental results by Doret et al. [3] - *a*, by DeMille et al. [1]- *b*, and by Fowler and Yellin [5]- *c*.

	$\alpha(6P_{1/2}) - \alpha(7S_{1/2})$	$\alpha(6P_{1/2}) - \alpha(7P_{1/2})$
Present work	-830	-4866
Expt.[a]	-829.7±3.1	
Expt.[b]	-900±48	-4967±249
Expt.[c]	-776±80	

experimental results from Refs. [1, 3, 5]. The conversion factor between the $\Delta\nu_S$ in kHz/(kV/cm)^2 units used in Refs. [1, 3, 5] to polarizabilities in atomic units used in the present work is $2\times10^{-7}h/(4\pi\epsilon_0 a_0^3) = 8.03756$, where h is the Plank constant. Our result for the $6P_{1/2}-7S_{1/2}$ Stark shift agrees with the most accurate, 0.4%, experimental value from Ref. [3] within the experimental uncertainty. The all-order value of the Stark shift for the $6P_{1/2}-7P_{1/2}$ transition also agrees with the experiment within the experimental uncertainty. We note that this comparison essentially tests the accuracy of the $7S$ and $7P_{1/2}$ polarizability calculations since the ground state polarizability is small comparing to the excited state polarizabilities.

IV. CONCLUSION

In summary, a systematic study using relativistic SD all-order method of the Stark shift within the $6P_{1/2}-7S_{1/2}$ and $6P_{1/2}-7P_{1/2}$ transitions and the Stark-induced amplitudes α_S and β_S in the $6P_{1/2}-7P_{1/2}$ transition in atomic thallium is presented. Our results $\alpha = 368$ a$_0{}^3$ and $|\beta_S|= 298$ a$_0{}^3$ support the experimental measurements carried out by D. DeMille, D. Budker, and E. D. Commins [Phys. Rev. A**50**, 4657 (1994)]. Our results for Stark shifts in the $6P_{1/2}-7S_{1/2}$ and $6P_{1/2}-7P_{1/2}$ transitions agree with the most accurate measured values within the experimental uncertainties for both transitions.

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