

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

M. S. Safronova\*

*Department of Physics and Astronomy, 223 Sharp Lab, University of Delaware, Newark, Delaware 19716, USA*

W. R. Johnson†

*Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556, USA*

U. I. Safronova‡ and T. E. Cowan

*Physics Department, University of Nevada, Reno, Nevada 89557, USA*

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Stark-induced amplitudes for the  $6P_{1/2}$ - $7P_{1/2}$  transition in Tl I are calculated using the relativistic singles-doubles (SD) approximation in which single and double excitations of Dirac-Hartree-Fock levels are summed to all orders in perturbation theory. Our SD values  $\alpha_S=368a_0^3$  and  $|\beta_S|=298a_0^3$  are in good agreement with the measurements  $\alpha_S=377(8)a_0^3$  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  $\beta_S$ . Measured values of both  $\alpha_S=2.01(4)\times 10^{-5}\mu_B$  cm/V and  $\beta_S=1.67(4)\times 10^{-5}\mu_B$  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  $\alpha_S=1.31(6)\times 10^{-5}\mu_B$  cm/V and  $\beta_S=1.09(5)\times 10^{-5}\mu_B$  cm/V by Tanner and Commins [2]. We note that in both experimental works, DeMille *et al.* [1], and Tanner and Commins [2],  $\alpha_S$  and  $\beta_S/\alpha_S$  have actually been measured, and the value of  $\beta_S$  was subsequently derived. The value for the ratio, measured by Tanner and Commins [2] was 0.83(1), while the value measured by DeMille *et al.* [1] was 0.85(3). However, the authors of Ref. [1] actually used the ratio value of Ref. [2], 0.83(1), to obtain the value of  $\beta_S$  as they believed it to be more accurate. In this work, we calculate the values of  $\alpha_S$  and  $\beta_S$  independently and obtain the theoretical value of the ratio. 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)<sup>2</sup> had higher accuracy by a 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)<sup>2</sup>.

In the present paper, we carry out relativistic all-order calculations of Stark-induced amplitudes  $\alpha_S$  and  $\beta_S$  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. Briefly, the wave function of the valence electron  $v$  is represented as an expansion

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

where  $\Phi_v$  is the lowest-order atomic state function, which is taken to be the *frozen-core* Dirac-Hartree-Fock (DHF) wave function of a state  $v$ . The indices  $m$  and  $n$  designate excited states and indices  $a$  and  $b$  designate core states. The equations for the excitation coefficients  $\rho_{ma}$ ,  $\rho_{mv}$ ,  $\rho_{mnab}$ , and  $\rho_{mnva}$  are solved iteratively until the correlation energy converges to the acceptable accuracy. The resulting values of the excitation coefficients are used to calculate matrix elements. We refer the reader to Refs. [6,7] for a detailed description of the approach. The resulting matrix elements contain the entire third-order many-body perturbation theory (MBPT) contribution as well as classes of fourth and higher orders of MBPT. Therefore, the SD all-order method is more accurate

\*Electronic address: [msafrono@udel.edu](mailto:msafrono@udel.edu)

†Electronic address: [johnson@nd.edu](mailto:johnson@nd.edu); URL: [www.nd.edu/~johnson](http://www.nd.edu/~johnson)

‡Electronic address: [usafrono@nd.edu](mailto:usafrono@nd.edu); On leave from ISAN, Troitsk, Russia

than the third-order MBPT. The advantage of the all-order method is that it treats correlations in a rather complete way yielding excellent results for the systems with one valence electron. We note that its implementation is entirely different from the configuration interaction method.

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 MBPT and the relativistic SD method. The SD calculations were found to be in excellent agreement with the best available experimental data. Our work [6] has established that all-order method is clearly suitable for the accurate calculation of TI properties.

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

Stark-induced scalar and vector polarizabilities  $\alpha_S$  and  $\beta_S$  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  $\alpha_S$  and  $\beta_S$  following the procedures used in [7] for alkali-metal atoms.

The scalar and vector polarizabilities  $\alpha_S$  and  $\beta_S$  for the transitions between the ground state  $6P_{1/2}$  and the excited state  $7P_{1/2}$  in Tl I are calculated as follows:

$$\alpha_S = \sum_n [I_\alpha(nS_{1/2}) - I_\alpha(nD_{3/2})], \quad (2)$$

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

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 follows [6]:

$$Z_{wv} = \frac{z_{wv} + Z^{(a)} + \dots + Z^{(t)}}{\sqrt{(1 + N_w)(1 + N_v)}}, \quad (4)$$

where  $z_{wv}$  is the lowest-order (DHF) matrix element and the terms  $Z^{(k)}$ ,  $k=a \dots t$  are linear or quadratic functions 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 fourth- and higher-order corrections.

The calculation of the  $\alpha_S$  is divided into three parts:

$$\alpha_S = \alpha_S^{\text{main}} + \alpha_S^{\text{tail}} + \delta\alpha_S^{\text{core}}, \quad (5)$$

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

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

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

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

where  $N$  is the number of the finite basis set states. The calculation of  $\beta_S$  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 present calculation of the polarizabilities required accurate representation of rather highly excited states, such as  $10S$  and  $9D$ , leading to the use of the large  $R=90$  a.u. cavity for the generation of the finite basis set and higher number of splines to produce high-accuracy single-particle orbitals. 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 interaction contributes about 1% to the values of  $\alpha_S$  and  $\beta_S$  at the DHF level. The BDHF approximation includes 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. (2) and (3) converge rapidly; therefore, only main term contributions have to be calculated accurately. We calculate main terms for  $\alpha_S$  and  $\beta_S$  using SD matrix elements and experimental energies [11]. The 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 the RPA approximation to evaluate the tail contribution for  $\alpha_S$  as DHF calculation significantly overestimates the tail yielding  $-25a_0^3$  while RPA calculation gives  $-16a_0^3$ . 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  $\alpha_S$  and is negligible at the present level of accuracy for  $\beta_S$ . It is evaluated in the DHF approximation.

In Tables I and II, we present the details of our  $\alpha_S$  and  $\beta_S$  calculations for the transition between the ground state  $6P_{1/2}$  and the excited  $7P_{1/2}$  state in Tl I. We separate the contribu-

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

Contribution	$nlj$	$E_{6P_{1/2}} - E_{nlj}$	$E_{7P_{1/2}} - E_{nlj}$	$Z_{7P_{1/2},nlj}$	$Z_{nlj,6P_{1/2}}$	$\alpha_S$
$\alpha_S^{\text{main}}(nS)$	$7S_{1/2}$	-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
$\alpha_S^{\text{tail}}(nS)$						-2.0
$\alpha_S^{\text{main}}(nD_{3/2})$	$6D_{3/2}$	-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
$\alpha_S^{\text{tail}}(nD_{3/2})$						-14.1
$\delta\alpha_S^{\text{core}}$						0.4
Total						368

tions from the  $nS$  and  $nD_{3/2}$  terms given by the Eqs. (2) and (3). Furthermore, the contribution from each  $n$  in the Eq. (6) for the main term is given separately in the corresponding ( $nlj$ ) row to demonstrate rapid convergence of the sums in Eqs. (2) and (3). 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  $\delta\alpha_S^{\text{core}}$  and  $\delta\beta_S^{\text{core}}$  of Tables I and II, respectively.

We list the values of the reduced electric-dipole matrix elements and energies used in the calculation of  $\alpha_S$  and  $\beta_S$  in Table I. Since the same matrix elements and energies contribute to  $\alpha_S$  and  $\beta_S$ , 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. (4)] are given in columns labeled  $Z_{wv}$ . It

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

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

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 a lesser extent, because of a 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  $\alpha_S$  and  $\beta_S$  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 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 procedure modifies the values of the  $7S$ - $7P_J$  and  $7P_{1/2}$ - $6D_{3/2}$  matrix elements by 2 and 3%, respectively. In systems with one valence electron, the scaling of the SD transition matrix values in similar cases generally produced the results that agreed with experimental values to 1% or better, even when the relative correlation correction was larger than for the present TI transitions. The scaling cannot be applied to improve the value of the  $6D_{3/2}$ - $6P_{1/2}$  matrix element as 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  $7P_{1/2}$ - $6D_{3/2}$ .

We find that the contribution of the  $6D_{3/2}$  term from Eqs. (2) and (3) dominates the values of  $\alpha_S$  and  $\beta_S$ . While all other valence terms contributing to  $\alpha_S$  have the same sign, there is significant cancellation of smaller terms contributing to  $\beta_S$ . In fact, all  $nS$  terms contribute with the sign opposite

TABLE III. The SD all-order Stark-induced scalar and vector polarizabilities for the  $6P_{1/2}$ - $7P_{1/2}$  transition Tl in are compared with measurements by Tanner and Commins [2] and DeMille *et al.* [1]. Units:  $a_0^3$ .

	$\alpha_S$	$ \beta_S $
This work	368	298
Expt. <sup>a</sup>	$247 \pm 12$	$198 \pm 10^b$
Expt. <sup>c</sup>	$377 \pm 8$	$313 \pm 8$

<sup>a</sup>Reference [2].

<sup>b</sup>This is a value directly quoted in  $a_0^3$  in Ref. [2]. If we recalculate the value  $\beta_S = 1.09(5) \times 10^{-5} \mu_B \text{ cm/V}$  quoted in the same paper to  $a_0^3$ , we obtain  $\beta_S = 205(10)a_0^3$ . Then, the resulting  $\beta_S/\alpha_S$  ratio is 0.83(1), as measured in Ref. [2].

<sup>c</sup>Reference [1].

to that of the  $nD_{3/2}$  terms with the exception of the first ones,  $7S$  and  $6D_{3/2}$ .

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  $(\mu_B/c)$  (cm/V) used in Refs. [1,2] and atomic units used in the present work is  $10^{-2} \alpha E_h / 2e a_0 = 1.8762 \times 10^7$ , 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  $\alpha_S$  is nearly within the experimental uncertainty of the Ref. [1] measurement and our value of  $\beta_S$  differs from the central experimental value of Ref. [1] by  $2\sigma$ . As we mentioned above, both DeMille *et al.* [1] and Tanner and Commins [2], has actually measured  $\alpha_S$  and  $\beta_S/\alpha_S$ . The authors of Ref. [1] actually used the ratio value of Ref. [2], 0.83(1), to obtain the value of  $\beta_S$  as they believed it to be more accurate. The theoretical value  $\beta_S/\alpha_S = 0.81$  is in good agreement with the experimental value 0.83(1).

### 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 differences of scalar dipole polarizabilities  $\alpha$  of the  $6P_{1/2}$  ground state and the  $7S_{1/2}$  or  $7P_{1/2}$  excited states. The expression for  $\alpha$  is given by (see, for example, Refs. [5,15]):

$$\alpha(n_0 P_{1/2}) = \sum_n [I_S(nS_{1/2}) + I_S(nD_{3/2})],$$

$$\alpha(n_0 S_{1/2}) = \sum_n [I_S(nP_{1/2}) + I_S(nP_{3/2})],$$

where

$$I_S(nlj) = \frac{1}{3} \frac{Z_{n_0 l_0 j_0, nlj}^2}{E_{nlj} - E_{n_0 l_0 j_0}}.$$

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 designa-

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

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

tions as in the previous section. The polarizability  $\alpha_{\text{core}}$  of the Hg-like ionic core is also evaluated using the RPA approximation. A more detailed discussion for the  $\alpha_{\text{core}}$  in Na, K, Rb, Cs, and Fr atomic systems is found in Ref. [7]. We note that contributions from  $\alpha_{\text{core}}$  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 the 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

TABLE V. The contributions to the scalar dipole  $7S$  polarizability  $\alpha$  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	$nlj$	$E_{nlj} - E_{7S}$	$ Z_{7S, nlj} $	$\alpha(7S_{1/2})$
$\alpha^{\text{main}}(nP_{1/2})$	$6P_{1/2}$	-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
$\alpha^{\text{tail}}(nP_{1/2})$				0.4
$\alpha^{\text{main}}(nP_{3/2})$	$6P_{3/2}$	-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
$\alpha^{\text{tail}}(nP_{3/2})$				3.0
$\alpha^{\text{core}}$				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], by DeMille *et al.* [1], and by Fowler and Yellin [5].

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

<sup>a</sup>Reference [3].

<sup>b</sup>Reference [1].

<sup>c</sup>Reference [5].

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 a more accurate evaluation of the dominant contributions to  $7S$  and  $7P_{1/2}$  polarizabilities. Our value for the  $\alpha(6P_{1/2})=50.4a_0^3$  is in good agreement with the theoretical result  $49.2a_0^3$  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 experimental results from Refs. [1,3,5]. The conversion factor between the  $\Delta\nu_S$  in kHz/(kV/cm)<sup>2</sup> units used in Refs. [1,3,5] to polarizabilities in atomic units used in the present work is  $2 \times 10^{-7}h/(4\pi\epsilon_0a_0^3)=8.03756$ , where  $h$  is the Planck 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.

Based on the analysis of the size and the breakdown of the correlation correction in this calculation, we expect our values to be accurate to a few percent. Higher accuracy is expected for  $7S$  and  $7P_{1/2}$  scalar dipole polarizabilities as in these cases all overwhelmingly dominant contributions ( $7S-7P_J$  and  $7P_{1/2}-6D_{3/2}$ ) contain estimates of the missing part of the correlation correction. The calculation of these two quantities is very much like the Ba<sup>+</sup> quadrupole polarizability where detailed uncertainty analysis yielded 2% accuracy. However, in TI SD all-order calculations, we are also dealing with the issue of attempting to describe a three-particle system as a one-particle system. The fact that we agree with the experiment on a number of TI quantities at the precision expected for such calculation for monovalent systems indicates that for these properties such an approximation does not cause significant additional error.

#### IV. CONCLUSION

In summary, a systematic study using the 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  $\alpha_S$  and  $\beta_S$  in the  $6P_{1/2}-7P_{1/2}$  transition in atomic thallium is presented. Our results,  $\alpha=368a_0^3$  and  $|\beta_S|=298a_0^3$ , 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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- [1] D. DeMille, D. Budker, and E. D. Commins, Phys. Rev. A **50**, 4657 (1994).  
 [2] C. E. Tanner and E. D. Commins, Phys. Rev. Lett. **56**, 332 (1986).  
 [3] S. C. Doret, P. D. Friedberg, A. J. Speck, D. S. Richardson, and P. K. Majumder, Phys. Rev. A **66**, 052504 (2002).  
 [4] P. A. Vetter, D. M. Meekhof, P. K. Majumder, S. K. Lamoreaux, and E. N. Fortson, Phys. Rev. Lett. **74**, 2658 (1995).  
 [5] T. R. Fowler and J. Yellin, Phys. Rev. A **1**, 1006 (1970).  
 [6] U. I. Safronova, M. S. Safronova, and W. R. Johnson, Phys. Rev. A **71**, 052506 (2005).  
 [7] M. S. Safronova, W. R. Johnson, and A. Derevianko, Phys. Rev. A **60**, 4476 (1999).  
 [8] W. R. Johnson, S. A. Blundell, and J. Sapirstein, Phys. Rev. A **37**, 307 (1988).  
 [9] A. Derevianko, Phys. Rev. Lett. **85**, 1618 (2000).  
 [10] A. Derevianko, Phys. Rev. A **65**, 012106 (2002).  
 [11] C. E. Moore, *Atomic Energy Levels—Vol. III, NSRDS-NBS 35* (U. S. Government Printing Office, Washington DC, 1971).  
 [12] S. A. Blundell, J. Sapirstein, and W. R. Johnson, Phys. Rev. D **45**, 1602 (1992).  
 [13] A. Kreuter, C. Becher, G. Lancaster, A. Mundt, C. Russo, H. Häffner, C. Roos, W. Hänsel, F. Schmidt-Kaler, R. Blatt *et al.*, Phys. Rev. A **71**, 032504 (2005).  
 [14] M. S. Safronova, Ph. D. thesis, Notre Dame University, 2000, (unpublished).  
 [15] M. S. Safronova and C. W. Clark, Phys. Rev. A **69**, 040501(R) (2004).  
 [16] M. G. Kozlov, S. G. Porsev, and W. R. Johnson, Phys. Rev. A **64**, 052107 (2001).