Polarizabilities, Stark shifts, and lifetimes of the In atom

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We evaluate the polarizabilities of the $5p_{1/2}$, $6s$, $6p_{1/2}$, and $6p_{3/2}$ states of In using two high-precision relativistic methods: the linearized coupled-cluster approach, where single, double, and partial triple excitations of the Dirac-Fock wave function are included to all orders of perturbation theory, and an approach that combines the configuration interaction and the coupled-cluster method. An extensive comparison of the accuracy of these methods is carried out. The uncertainties of all recommended values are evaluated. Our result for the $6s$-$5p_{1/2}$ Stark shift is in excellent agreement with a recent measurement [Ranjit *et al.*, Phys. Rev. A **87**, 032506 (2013)]. Combining our calculation with this precision measurement allows us to infer the values of the $6p_{1/2}$ and $6p_{3/2}$ lifetimes in In with 0.8% accuracy. Our predictions for the $6p_{3/2}$ scalar and tensor polarizabilities may be combined with the future measurement of the $6s$-$6p_{3/2}$ Stark shift to accurately determine the $5d_j$-$6p_{j'}$ matrix elements.

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I. INTRODUCTION

An indium atom represents an excellent system to compare the accuracy of different high-precision theoretical methods since it may be considered both a monovalent system (assuming a closed $5s^2$ core) and a trivalent system. In this work, we use both the linearized coupled-cluster (CC) approach, where single, double, and partial triple excitations (LCCSDpT) of the Dirac-Fock wave function are included to all orders, and a method combining the configuration interaction (CI) and the CC method (CI + all order).

The monovalent LCCSDpT method has been applied to a large number of neutral and ionized monovalent systems and yielded very accurate predictions for a number of atomic properties (see review [1] and references therein). It has been used for a variety of applications, ranging from the study of fundamental symmetries [2,3] to the study of degenerate quantum gases and quantum information [4]. This method has been tested against all high-precision alkali and monovalent ion experimental values, which allowed us to establish a systematic procedure to evaluate its uncertainties even when no experimental data exist [5].

The CI + all-order method was recently developed for the treatment of more complicated systems [6]. It has been tested on a variety of divalent systems [7–11] and applied to Tl [12]. However, there are far fewer experimental benchmarks on the properties of divalent and trivalent systems in comparison with alkalis. A recent high-precision (0.27%) measurement of the $6s$-$5p_{1/2}$ Stark shift in In [13] provides an excellent opportunity to compare the accuracy of the CI + all-order and LCCSDpT approaches and test the procedure for evaluation of the uncertainties of theoretical data.

In turn, the combination of the Stark shift measurement [13] and our calculations allows us to infer the values of the $6p_{1/2}$ and $6p_{3/2}$ lifetimes in In with 0.8% accuracy. It is extremely difficult to achieve such a high accuracy via direct lifetime measurements. Moreover, we predict the values of the $6p_{3/2}$ scalar and tensor polarizabilities, which can soon be tested against the future measurement of the $6s$-$6p_{3/2}$ Stark shift [13] and will allow to determine the $5d$-$6p$ matrix elements.

Precise knowledge of In properties is of interest for the study of fundamental symmetries, including parity violation and the search for the permanent electric-dipole moment, since it is similar to Tl (also group IIIB). Understanding of the theoretical and experimental uncertainties in In can be used in Tl studies. Both electric dipole moment [14] and parity-violation studies [15,16] had been carried out in Tl. The theory accuracy in Tl is still below the experimental accuracy, hindering further parity violation studies using this system. A recent controversy regarding the calculated values of the Tl electric dipole moment enhancement factor is discussed in detail in [12].

In-like ions are excellent candidates for the search for variation of the fine-structure constant α. Despite very high ionization energies, certain ions have transitions that lie in the optical range due to level crossing and are very sensitive to α variation [17]. In-like ions are particulary well suited for the experimental search for such transitions [18]. In fact, an In-like isoelectronic sequence has by far the largest number of ions with long-lived metastable states, transitions with wavelengths between 170 and 3000 nm and high sensitivity to α variation, and stable isotopes [18]. One of the main obstacles for experimental work in this direction is the lack of any experimental data for these systems and the difficulty of accurate theoretical predictions. Testing the CI + all-order method on neutral In provides important information on the accuracy of this approach for further studies of In-like ions.

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II. METHOD

We use two relativistic high-precision approaches for all calculations in this work. The first method (LCCSDpT) has been extremely successful in predicting properties of alkali-metal atoms and other monovalent ions [1]. It can also be applied to In by treating the $5s^2$ shell as a part of the $[1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 5s^2]$ core. The disadvantage of this approach is its inability to explicitly treat three-particle states, such as $5s5p^2$. However, the LCCSDpT method produced results for the $6p_{1/2}$-$7s$ and $6p_{1/2}$-$7p_{1/2}$ Stark shifts [19] of Tl, which is a similar group IIIB system, in excellent agreement with experiments [20,21]. For convenience, we refer to the LCCSDpT approach as the CC method in the text and tables below.

The CI + all-order method (we also refer to it as CI + All in the text and tables below) was developed in [6]. It allows us to treat In as a three-particle system, so all three electrons above the $4d^{10}$ shell are considered valence. In this approach, the CC method is first used to accurately describe core-core and core-valence correlation and to incorporate them into the effective Hamiltonian. Therefore, the core-core and core-valence sectors of the correlation corrections for systems with few valence electrons are treated with the same accuracy as in the all-order approach for monovalent atoms. Then the CI method is used to treat valence-valence correlations. Since the CI space includes only three electrons, it can be made essentially complete. This method allows us to include correlation corrections to the wave functions in a more complete way than the CC approach does. In particular, it is capable of accurately accounting for the configuration mixing.

The CI + all-order method yielded accurate wave functions for calculations of atomic properties, such as lifetimes, polarizabilities, and hyperfine-structure constants, for a number of divalent systems and Tl [6–12]. However, the various types of corrections to the effective dipole operator $D_{\rm eff}$ are included in a more complete way in the CC approach at present. Therefore, both approaches are complementary and the difference in the results can serve as an estimate of the uncertainties.

First, we compare the In energy levels calculated using the CC and CI + all-order methods with experiment [22] in Table I. The CC values include the part of the third-order energy not included by the LCCSD method and the Breit interaction to second order. The CI + all-order method includes the Breit interaction on the same footing as the Coulomb interaction, which would include some higher order Breit corrections. The Breit correction is small; however, it significantly improves the accuracy of the $5s5p^2 \, ^4P_J$ triplet splitting. Both calculations are carried out with $l_{\rm max} = 6$ partial waves in all intermediate sums in the many-body expressions and include extrapolation for the contributions of partial waves with $l > 6$.

In CC calculations, extrapolation is carried out to second order using a separate code. This second-order calculation shows that the total contribution of $l > 6$ partial waves is close to the contribution of the single $l = 6$ partial wave. This empirical observation is used to estimate the contribution of higher order partial waves in the CI + all-order approach. We find that while both methods give energy levels in very good agreement with experiment, the CI + all-order

TABLE I. Comparison of the CC and CI + all-order (CI + All) energies of In levels with experiment [22]. Three-electron binding energies are given in the first row. Energies in other rows are given relative to the ground state. Corresponding relative differences of these two calculations from experiment are given in the column labeled "Diff." The $5s^2 6p \, ^2P_{1/2}^o$-$5s^2 6s \, ^2S_{1/2}$ and $5s^2 5d \, ^2D_{5/2}$-$5s^2 6p \, ^2P_{3/2}^o$ transition energies are listed in the last row.

State		Expt.	CC	Diff.	CI + All	Diff.
$5s^2 5p$	$^2P_{1/2}^o$	425060			425719	0.15%
	$^2P_{3/2}^o$	2213	2168	2.02%	2195	0.82%
$5s^2 6p$	$^2P_{1/2}^o$	31817	31468	1.10%	31805	0.04%
	$^2P_{3/2}^o$	32115	31769	1.08%	32104	0.03%
$5s^2 7p$	$^2P_{1/2}^o$	38861	38513	0.90%	38911	-0.13%
	$^2P_{3/2}^o$	38973	38625	0.89%	39023	-0.13%
$5s^2 6s$	$^2S_{1/2}$	24373	23862	2.10%	24272	0.41%
$5s^2 5d$	$^2D_{3/2}$	32892	32563	1.00%	32836	0.17%
	$^2D_{5/2}$	32916	32754	0.49%	32863	0.16%
$5s5p^2$	$^4P_{1/2}$	34978			35299	-0.92%
	$^4P_{3/2}$	36021			36346	-0.90%
	$^4P_{5/2}$	37452			37770	-0.85%
$5s^2 7s$	$^2S_{1/2}$	36302	35928	1.03%	36284	0.05%
$\Delta(6p_{1/2}$-$6s)$		7444	7606	-2.18%	7531	-1.16%
$\Delta(5d_{5/2}$-$6p_{3/2})$		800	985	-23%	759	5.16%

results systematically agree better with the experimental values.

III. POLARIZABILITIES

The static polarizability of a state with total angular momentum J, its projection M, and energy E can be expressed as a sum over unperturbed intermediate states,

$$\alpha(J,M) = 2 \sum_n \frac{|\langle J,M|D_z|J_n,M\rangle|^2}{E_n - E}, \qquad (1)$$

where J_n and E_n are the total angular momenta and the energies of the intermediate states.

The static polarizability $\alpha(J,M)$ can be conveniently decomposed into scalar and tensor parts according to

$$\alpha = \alpha_0 + \alpha_2 \frac{3M^2 - J(J+1)}{J(2J-1)}, \qquad (2)$$

where the scalar (α_0) and tensor (α_2) polarizabilities can be expressed as [23]

$$\alpha_0 = \frac{2}{3(2J+1)} \sum_n \frac{|\langle J||D||J_n\rangle|^2}{E_n - E} \qquad (3)$$

and

$$\alpha_2 = 4\left(\frac{5J(2J-1)}{6(2J+3)(2J+1)(J+1)}\right)^{1/2}$$
$$\times \sum_n (-1)^{J+J_n} \begin{Bmatrix} J & 1 & J_n \\ 1 & J & 2 \end{Bmatrix} \frac{|\langle J||D||J_n\rangle|^2}{E_n - E}. \qquad (4)$$

For an open-shell atom, α_0 may be separated into a contribution from the valence electrons, α_0^v, a contribution from the core electrons, α^c, and a core modification due to the presence of the valence electrons, α^{vc}. We calculate the core

and α^{vc} terms using the random phase approximation (RPA) in both approaches. The valence parts are calculated differently in the CC and CI + all-order methods.

In the CC approach, the valence polarizability of the single-electron valence state $|w\rangle$ is calculated using the sum-over-states method,

$$\alpha_0^v = \frac{2}{3(2j_w + 1)} \sum_k \frac{\langle k\|D\|w\rangle^2}{E_k - E_w},\qquad(5)$$

where j_w is the total angular momentum of state $|w\rangle$ and $\langle k\|D\|w\rangle$ are the single-electron reduced electric-dipole matrix elements and sum over k runs over all intermediate excited states allowed by the electric-dipole transition rules [24].

In the CI + all-order approach, the valence part of the polarizability is determined by solving the inhomogeneous equation of perturbation theory in the valence space, which is approximated as [25]

$$(E - H_{\text{eff}})|\gamma, M'\rangle = (D_{\text{eff}})_q|\gamma, J, M\rangle,\qquad(6)$$

where γ incorporates all other quantum numbers except J and M. The wave function $|\gamma, M'\rangle$, where $M' = M + q$, is composed of parts that have angular momenta of $J' = J, J \pm 1$. The construction of the effective Hamiltonian H_{eff} using the all-order approach is described in [6]. The effective dipole operator D_{eff} includes RPA corrections.

While we do not use the sum-over-state approach in the calculations of the polarizabilities in the CI + all-order method, it is useful to calculate several dominant contributions to polarizabilities by combining the CI + all-order values of the $E1$ matrix elements and energies according to the sum-over-states formula (5). This allows us to compare dominant terms and total remainders calculated by the CC and CI + all-order methods.

We compare the contributions to the $6s$ and $5p_{1/2}$ polarizabilities α_0 calculated by two approaches in Table II. The CC and CI + all-order reduced electric-dipole matrix elements and the contributions to the polarizability α_0 are listed in the columns labeled "CC" and "CI + All," respectively. The experimental energies listed in column "ΔE" are used to calculate the dominant contributions for consistency and to improve the accuracy. We find generally very good agreement of the CC and CI + all-order results, with the exception of the $5p_{1/2}$-$6d_{3/2}$ and the $5p_{1/2}$-$7d_{3/2}$ cases. These matrix elements are small and have very large correlation corrections. The uncertainties in the CC contributions are evaluated using the well-defined approach descried in detail in [5]. It involves calculation of the spread of four different CC calculations of increasing accuracy for each matrix element. The results labeled "Other" in the "CI + All" column are obtained by subtracting the separately listed dominant terms from the final valence value.

We note that the core contribution in the CC approach is substantially larger then that in the CI + all-order approach since the $5s$ shell is included in the core in the CC method but not in the CI + all-order one. The uncertainty of the CC core $+ vc$ term is evaluated as the difference of the Dirac-Hartree-Fock (DHF) and RPA total core $+ vc$ values. The uncertainties in the tail are determined based on the difference in the RPA versus all-order values for terms with $n = 9$–12.

TABLE II. Contributions to the $6s$ and $5p_{1/2}$ static polarizabilities are listed (in a_0^3) in columns labeled "α_0." The experimental energies (in cm^{-1}) and the theoretical electric-dipole reduced matrix elements (in a.u.) used to calculate the dominant contributions are listed in the columns labeled "ΔE" and "D". The CC and CI + all-order electric-dipole matrix elements and the polarizability contributions are listed in the columns labeled "CC" and "CI + All," respectively.

Contribution	ΔE Expt.	D CC	D CI + All	α_0 CC	α_0 CI + All
			$6s$ polarizability		
$5p_{1/2}$	-24373	1.911	1.885	$-11.0(6)$	-10.7
$6p_{1/2}$	7444	6.110	6.081	367(12)	364
$7p_{1/2}$	14488	0.683	0.648	2.4(4)	2.1
$8p_{1/2}$	17454	0.277	0.265	0.3(1)	0.3
$(9\text{–}12)p_{1/2}$				0.17(3)	
$(n > 12)p_{1/2}$				0.14(5)	
$5p_{3/2}$	-22160	2.935	2.899	$-28(1)$	-28
$6p_{3/2}$	7742	8.529	8.491	687(24)	681
$7p_{3/2}$	14600	1.131	1.084	6.4(1.0)	5.9
$8p_{3/2}$	17508	0.489	0.479	1.0(2)	1.0
$(9\text{–}12)p_{3/2}$				0.6(1)	
$(n > 12)p_{1/2}$				0.6(2)	
Other					23
Core				29.6(4.0)	3.2
vc				0.0	0.0
Main				1024.9	1015.5
Remainder				31.1	26.2
Total				1056(27)	1042
			$5p_{1/2}$ polarizability		
$6s$	24373	1.911	1.885	11.0(6)	10.7
$7s$	36302	0.548	0.534	0.61(3)	0.57
$8s$	40637	0.297	0.297	0.16(1)	0.16
$(9\text{–}12)s$				0.13(3)	
$(n > 12)s$				0.7(2)	
$5d_{3/2}$	32892	2.623	2.577	15.3(1.5)	14.8
$6d_{3/2}$	39049	1.001	0.865	1.9(2)	1.40
$7d_{3/2}$	41836	0.537	0.308	0.5(1)	0.17
$(8\text{–}12)d_{3/2}$				0.6(2)	
$(n > 12)d_{3/2}$				6.1(4.0)	
Other					31.6
Core[a]				29.6(2.5)	3.22
vc				-5.0	-0.15
Main				29.4	27.7
Remainder				32.1	34.6
Total				61.5(5.6)	62.4

[a]The uncertainty is the sum of the core and vc uncertainties.

As an additional comparison between the two approaches, we group eight contributions to the $6s$ polarizability [$6s - (5\text{–}8)p_{1/2,3/2}$] together and list them in the row "Main." The contributions from six terms ($6s$, $7s$, $8s$, $5d_{3/2}$, $6d_{3/2}$, and $7d_{3/2}$) are grouped together in the row "Main" for the $5p_{1/2}$ polarizability. These main terms are subtracted from the totals to obtain the contributions from the remaining terms. These terms are listed in the rows labeled "Remainder." The difference of the remainder part for the $6s$ polarizability appears to indicate a lower value of the core polarizability when $5s^2$ is included in the core. Overall, there is good agreement of both the main and the remainder parts between

TABLE III. Contributions to the $6p_{1/2}$ static polarizability are listed (in a_0^3) in columns labeled "α_0". The experimental energies (in cm^{-1}) and the theoretical electric-dipole reduced matrix elements (in a.u.) used to calculate the dominant contributions are listed in the columns labeled "ΔE" and "D". The CC and CI + all-order matrix elements and the polarizability contributions are listed in the columns labeled "CC" and "CI + All," respectively. The contributions from the terms $6s$, $7s$, $8s$, $5d_{3/2}$, $6d_{3/2}$, and $7d_{3/2}$ are grouped together in the row "Main".

Contribution	ΔE Expt.	D CC	D CI + All	α_0 CC	α_0 CI + All
$6s$	−7444	6.110	6.081	−367(12)	−363
$7s$	4485	6.289	6.239	645(5)	635
$8s$	8820	1.294	1.317	14	14
$(9–12)s$				5(1)	
$(n > 12)s$				4(1)	
$5d_{3/2}$	1075	10.095	9.893	6933(140)	6659
$6d_{3/2}$	7232	6.470	6.477	423(65)	424
$7d_{3/2}$	10019	2.861	2.848	60(7)	59
$(8–12)d_{3/2}$				35(11)	
$(n > 12)d_{3/2}$				35(6)	
Other					81
Core				30(4)	3.2
Main				7709(154)	7429
Remainder				108(13)	84
Total				7817(155)	7513
Recommended				7817(300)	

the two calculations. The final values for the $6s$ and $5p_{1/2}$ polarizabilities, the $6s$-$5p_{1/2}$ Stark shift, and their uncertainties are discussed in the next section.

Contributions to the $6p_{1/2}$ and $6p_{3/2}$ polarizabilities of In (in a_0^3) are listed in Tables III and IV, respectively. The tensor polarizability α_2 of the $6p_{3/2}$ state is given in Table IV. These tables are structured in exactly the same way as Table II. The results of both CC and CI + all-order calculations are given. The only difference is the listing of the $5s5p^2\,^4P_{5/2}$ contribution to the $6p_{3/2}$ polarizability. The contributions of the $5s5p^2\,^4P_J$ state to all other polarizabilities considered here were found to be negligible.

We find a 3.9% difference between the CC and the CI + all-order values for both scalar $6p_j$ polarizabilities and a 3.1% difference between the values of the tensor α_2 polarizability for the $6p_{3/2}$ state. These differences are caused by the 2% difference in the values of the $6p_{1/2}$-$5d_{3/2}$, $6p_{3/2}$-$5d_{3/2}$, and $6p_{3/2}$-$5d_{5/2}$ matrix elements which dominate the $6p_j$ polarizability values. This 2% difference is consistent with the expected accuracy of the CI + all-order method for these transitions. We evaluate the accuracy of the CC values to be of the order of 1%. The uncertainty evaluation is carried out differently for these three transitions owing to a convergence problem of the all-order equations for $5d$ states. We performed the calculations with three and five iterations in the LCCSD approximation and in the LCCSDpT approximation and carried out the scaling procedure (described, for example, in Ref. [1]) using these four different starting points. The spread of the resulting scaled values is 0.7%. Since the scaling estimates the dominant omitted corrections in such

transitions, all other omitted corrections should not exceed this upper bound of 0.7%, resulting in a total uncertainty estimate of 1%. The CC and CI + all-order values for all other contributions, including small remainders, were found to be in good agreement. Because we expect the CC method to yield more accurate values of the $6s$-$5d$ matrix elements, we take the CC values of the $6p$ polarizabilities as the final results.

Most likely, the discrepancy with the CI + all-order calculations is caused by an omission of the small corrections to the effective dipole operator in the CI + all-order approach. However, it might be possible that the $6p$-$5d$ matrix elements are affected by the small mixing of the even $5s^25d$ states with the $5s5p^2\,^4P_J$ triplet, which is accounted for by the CI + all-order method but not the CC method. The weight (in probability) of the $5s5p^2$ configuration in the $5d_j$ levels is 0.02–0.03, i.e., small but non-negligible. Moreover, there is a very strong mixing of the nd configurations. Therefore, we take the difference between the CI + all-order and the CC results as the final uncertainty to account for the possible uncertainty due to such mixing. We note that the $5s^26s$ state essentially does not mix with the $5s5p^2$ configuration (its weight is only 0.0003), so $6s$-$6p$ matrix elements are not affected by such mixing. A high-precision measurement of the $6p_{3/2}$-$6s$ Stark shift should resolve this question. The final values for the $6p_{3/2}$ polarizabilities are listed in the last rows in Tables III and IV.

IV. DISCUSSION OF THE RESULTS AND THEIR UNCERTAINTIES

While we have estimated the uncertainties of the CC calculation, it is possible to improve our evaluation of the uncertainties by comparing the CC and CI + all-order results. As we have described above, these two high-precision approaches include somewhat different higher order effects. The CI + all-order calculations include the valence-valence correlation corrections to the wave functions very precisely, as indicated by the excellent agreement of the respective energies with experiment. On the other hand, the effective dipole operator $D_{\rm eff}$ includes only RPA corrections in the CI + all-order method at the present time, omitting the structure radiation, the normalization, and other small corrections. These corrections to the electric-dipole matrix elements are included in the CC method.

In the polarizability calculations, we use the experimental energies for the main terms. Therefore, a more accurate method of calculating individual matrix elements is somewhat more important for the $6s$ polarizability. In the framework of the CC method, the $5s$ shell is included in the core and we have a large core polarizability. It leads to an increase in the total uncertainty of the polarizability because the core polarizability is calculated with less accuracy. But it cancels out when the Stark shift of a transition is calculated. The CI + all-order method treats contributions with a high n with a better accuracy, since it is done by solving the inhomogeneous equation instead of using the sum-over-states method. This is not significant for the $6s$ polarizability, where such tail contributions are small, but is important for the $5p_{1/2}$ polarizability.

In summary, the CC and CI + all-order methods together include all correlation corrections that are expected to be

TABLE IV. Contributions to the $6p_{3/2}$ scalar and tensor polarizabilities are listed (in a_0^3) in the columns labeled "α_0" and "α_2". The experimental energies (in cm^{-1}) and the theoretical electric-dipole reduced matrix elements (in a.u.) used to calculate the dominant contributions are listed in the columns labeled "ΔE" and "D". The CC and CI + all-order matrix elements and the scalar and tensor polarizability contributions are listed in the columns labeled "CC" and "CI + All", respectively. The contributions from the terms $6s$, $7s$, $8s$, $5d_j$, $6d_j$, and $7d_j$ are grouped together in the row "Main".

	ΔE	D		α_0		α_2	
Contribution	Expt.	CC	CI + All	CC	CI + All	CC	CI + All
$6s$	−7742	8.529	8.491	−344(12)	−341	344(12)	341
$7s$	4187	9.413	9.338	774(7)	762	−774(7)	−762
$8s$	8522	1.797	1.830	14	14	−14	−14
$(9–12)s$				5		−5	
$(n > 12)s$				4(1)		−4(1)	
$5d_{3/2}$	777	4.511	4.420	958(19)	920	767(15)	736
$6d_{3/2}$	6933	3.157	3.124	53(7)	51	42(6)	41
$7d_{3/2}$	9721	1.350	1.309	7(1)	6	5(1)	5
$(8–12)d_{3/2}$				4(1)		3(1)	
$(n > 12)d_{3/2}$				4(1)		3(1)	
$5d_{5/2}$	800	13.577	13.296	8426(170)	8080	−1685(34)	−1616
$6d_{5/2}$	6983	9.218	9.031	445(58)	427	−89(12)	−85
$7d_{5/2}$	9747	4.003	3.930	60(7)	58	−12(1)	−12
$(8–12)d_{5/2}$				35(7)		−7(1)	
$(n > 12)d_{5/2}$				33(9)		−7(1)	
$5s5p^2\ ^4P_{5/2}$	5337		2.337		37		−7
Other					79		−13
Core				30(4)	3		
Main				10393(180)	9979	−1416(42)	−1367
Remainder				114(12)	119	−16(2)	−20
Total				10506(180)	10098	−1432(42)	−1387
Recommended				10500(400)		−1432(45)	

dominant for the present polarizability calculations. Therefore, the uncertainty can be approximated as the difference $\delta\alpha = |\alpha(\text{CC}) - \alpha(\text{CI} + \text{all})|$ of the CC and CI + all-order results. All other omitted higher order corrections are expected to be smaller than already included ones; therefore, we can also assume that they do not exceed $\delta\alpha$. Therefore, we take our final uncertainty in the polarizabilities and their difference to be $\sqrt{2}\delta\alpha$, calculated separately for each of the properties. We list our final values of the $6s$ and $5p_{1/2}$ polarizabilities and their difference $\Delta\alpha_0(6s\text{-}5p_{1/2})$ in Table V (in a.u.).

We take the CC results to be the final values since the CC method accounts for more higher order corrections to

the $E1$ matrix elements that dominate the $6s$ polarizability. For consistency, the CC value of the $5p_{1/2}$ polarizability is used when calculating the final value for the $6s$-$5p_{1/2}$ Stark shift. Our final result is in excellent agreement with a recent high-precision measurement of the $6s$-$5p_{1/2}$ Stark shift [13], which allows for a benchmark comparison of the theoretical approaches. We find that the CC value is closer to the experimental measurement than the CI + all-order result. Our calculated polarizability of the $5p_{1/2}$ state is in excellent agreement with recent CC single, double, and perturbative triple excitation [CCSD(T)] calculations in Refs. [26] and [27]. We note that our implementation of the CC method differs

TABLE V. Final values of the $6s$ and $5p_{1/2}$ polarizabilities and their difference $\Delta\alpha_0$ (in a.u.). Determination of the reduced electric-dipole $6s$-$6p_j$ matrix elements (in a.u.) and $6p_j$ lifetimes (in ns) from the combination of the recently measured Stark shift [13] and theoretical values. The quantity C is the value of $\Delta\alpha_0(6s\text{-}5p_{1/2})$ with the contribution of $6s$-$6p_j$ transitions subtracted out. The results are compared with other theory and experiment.

	$\alpha_0(6s)$	$\alpha_0(5p_{1/2})$	$\Delta\alpha_0(6s\text{-}5p_{1/2})$	C	$D(6s\text{-}6p_{1/2})$	$D(6s\text{-}6p_{3/2})$	$\tau(6p_{1/2})$	$\tau(6p_{3/2})$
CC	1056	61.5	995	−59.6	6.126	8.551		
CI + All	1042	62.4	980	−65.2	6.141	8.575		
Final	1056(20)	61.5(1.3)	995(21)	−59.6(7.8)	6.126(24)	8.551(34)	63.77(50)	58.17(45)
Expt. [13]			1000.2(2.7)					
Expt. [28]		69(8)						
Theory [27]		61.48						
Theory [26]		62.0(1.9)						
Theory [29]							63.8(8)	58(1)

significantly from that in Refs. [26] and [27] (see the recent review [24] for details).

V. DETERMINATION OF LIFETIMES

Recent precision measurement of the $6s$-$5p_{1/2}$ Stark shift [13] can be combined with the present calculations to obtain very accurate lifetimes of the $6p_{1/2}$ and $6p_{3/2}$ states. This is possible since the $6s$-$6p_j$ matrix elements overwhelmingly dominate the values of this Stark shift, as illustrated by Table II. Separating the $6s$-$6p_j$ contributions [see Eq. (5)], we write the $\Delta\alpha_0(6s$-$5p_{1/2})$ Stark shift as

$$\Delta\alpha_0(6s - 5p_{1/2}) = BS + C, \quad (7)$$

where

$$B = \frac{1}{3}\left(\frac{1}{E(6p_{1/2}) - E(6s)} + \frac{R^2}{E(6p_{3/2}) - E(6s)}\right), \quad (8)$$

$S = D^2$ is the $6s$-$6p_{1/2}$ line strength, R is the ratio of the $D(6s$-$6p_{3/2})$ and $D(6s$-$6p_{1/2})$ reduced $E1$ matrix elements, and the term C contains all other contributions to the Stark shift. We calculate the ratio R to be 1.396(1). The uncertainty (0.001) is very small since the ratio R is very insensitive to different corrections. Using the results presented in Table II, we see that the CC and CI + all-order methods give R values equal to each other up to the fourth significant figure. Substituting the ratio R and the corresponding experimental energies in Eq. (8) gives $B = 28.24(3)$ a.u.

The values of C in the CC and CI + all-order methods are obtained from the results presented in Table II and are equal to -59.6 and -65.2 a.u., respectively (see Table V). Adding the relevant uncertainties from CC calculations in Table II in quadrature leads to the uncertainty $\delta C = 4.9$ a.u. This number is consistent with the difference in the CC and CI + all-order values, 5.6 a.u. We note that the uncertainty of the core term does not contribute to δC, since this term is canceled when the Stark shift is calculated. The $5p_{1/2}$ α_{vc} term does contribute, and its uncertainty is 1.3 a.u. To evaluate the final uncertainty in C we use the same rule as for the polarizability: multiply the difference in the CC and CI + all-order values by $\sqrt{2}$ to account for other small uncertainties not included in our consideration. Again, we assume that they cannot exceed the difference between the CC and the CI + all-order values. The final value for C is presented in Table V.

There are three sources of the uncertainties contributing to the uncertainty in $D(6s$-$6p_{1/2})$: uncertainties in C, B, and experimental values of $\Delta\alpha_0$. For convenience, we calculate first the uncertainty in the line strength S using the formula

$$\delta S = \frac{1}{B}\sqrt{(\delta C)^2 + (\delta\Delta\alpha_0)^2 + (S\delta B)^2}. \quad (9)$$

The relative uncertainty in D is one-half of the relative uncertainty in S. The uncertainty in B is negligible. The final values of the matrix elements are listed in Table V. The lifetimes of the $6p_{1/2}$ and $6p_{3/2}$ states are obtained using the formula $\tau_a = 1/A_{ab}$ since there is only one $E1$ decay channel for each state. The transition rate A_{ab} is given by

$$A_{ab} = \frac{2.02613 \times 10^{18}}{\lambda_{ab}^3}\frac{S_{ab}}{2J_a + 1} \text{ s}^{-1}, \quad (10)$$

where the transition wavelength λ_{ab} is in Å. The relative uncertainties in the lifetimes are twice that of the relative uncertainties of the corresponding $E1$ matrix elements. The final values are listed in Table V.

To simplify the extraction of the $5d$-$6p$ matrix elements from the future experimental value of the $6p_{3/2}$-$6s$ Stark shift, we evaluated the sum of all contributions to the $6p_{1/2}$ and $6p_{3/2}$ polarizabilities with the $5d$-$6p$ terms excluded. These quantities, which we designate $\widetilde{C}(6p_j)$, are obtained from the data in Tables III and IV. We note that $\widetilde{C}(6p_{1/2})$ and $\widetilde{C}(6p_{3/2})$ refer to the contributions to the polarizabilities, rather than their differences as in Eq. (7).

We find that the CC and CI + all-order results are very close together and are well within the uncertainty estimates of the CC data. The CC values (in a.u.) are $\widetilde{C}(6p_{1/2}) = 884(68)$, $\widetilde{C}_0(6p_{3/2}) = 1123(60)$, and $\widetilde{C}_2(6p_{3/2}) = -514(19)$, where subscripts "0" and "2" refer to the scalar and tensor polarizabilities, respectively. The corresponding CI + all-order values are 854, 1098, and -506 (in a.u.). The ratio of the $6p_{1/2}$-$5d_{3/2}$ and $6p_{3/2}$-$5d_{3/2}$ matrix elements is 2.238(4) and the ratio of the $6p_{3/2}$-$5d_{5/2}$ and $6p_{3/2}$-$5d_{3/2}$ matrix elements is 3.0095(16).

VI. CONCLUSION

We have carried out a systematic comparison of the linearized CC and CI + all-order method using the polarizabilities of the low-lying states of the In atom as a benchmark testing case. We find that the CI + all-order method produces more accurate data for the energy levels. It appears that the CC data for the $E1$ matrix elements are somewhat more accurate due to the more complete inclusion of the small higher order corrections to the matrix elements in cases where relevant configuration mixing of trivalent states is negligible. This is an additional motivation for incorporating such corrections into the CI + all-order formalism at the all-order level in the future.

Our result for the $6s$-$5p_{1/2}$ Stark shift is in excellent agreement with the recent high-precision experiment [13]. We also provide predictions for the polarizabilities of the $6p_{1/2}$ and $6p_{3/2}$ states. A precise experimental measurement of the $6p_{3/2}$-$6s$ Stark shift proposed in [13] would be a good test of our calculations. This will also provide an excellent test of the theoretical approaches. Combining the present calculations with the experimental Stark shift data allows very accurate extraction of the lifetimes of low-lying In states.

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