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Inconsistencies between lifetime and polarizability measurements in Cs

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Electric dipole matrix elements for 6*p*-*nd*, $n=5$, 6, 7, transitions in cesium are calculated using a relativistic all-order method. The resulting matrix elements are used to evaluate 5*d* lifetimes and 6*p* polarizabilities. The data are compared with experimental lifetime and polarizability measurements made by different groups. Domination of the 6*p* scalar polarizabilities by 5*d*-6*p* dipole matrix elements facilitates an exacting consistency check of 5*d* lifetime and 6*p* polarizability data. Values of 5*d*-6*p* matrix elements obtained from experimental 5*d* lifetime data are found to be inconsistent with those inferred from 6*p* polarizabilities derived from experimental Stark shift data. Our *ab initio* calculated 6*p* polarizabilities agree well with experimental determinations.

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The understanding of the accuracy of *ab initio* calculations in cesium is vital for the analysis of the Cs parity nonconservation (PNC) experiment [1]. In 1999, motivated by a number of recent high-precision experiments, Bennett and Wieman [2] reanalyzed the agreement of theoretical calculations and experimental data for a number of Cs atomic properties and reduced the previous theoretical uncertainty in the PNC amplitude by a factor of 2. Utilizing measurements of the tensor transition polarizability β reported in same work, they demonstrated a 2.5σ discrepancy between the value of the weak charge Q_W predicted by the standard model and that derived from the Cs PNC experiment. Although several papers (for example, Refs. [3–9]) have addressed this disagreement since 1999, the issue of the accuracy of *ab initio* calculations in Cs continues to be of interest.

In this work, we investigate the radiative properties of Cs 6*p*-*nd* transitions. Although these do not bear directly on PNC experiments done to date, they have been the subject of careful experimental investigation and thus provide benchmarks for precise comparison of theory and experiment. In particular, there exist two independent measurements of the lifetimes of the 5*d* states [10,11], which do not agree within their stated uncertainties. There also exist several experimental determinations of the 6*p*-6*s* Stark shifts which allow one to infer the values of polarizabilities of the 6*p* states [12–14]. Here we show that *ab initio* theory can check the mutual consistency of 5*d* lifetime and 6*p* polarizability data, with an accuracy of about 1%. We find the lifetime and polarizability results to be inconsistent at this level. Our calculations agree with the experimental values of 6*p* polarizabilities, but deviate from both determinations of the 5*d* lifetimes. We suggest that further experiments are desirable in order to clarify this issue. In addition, understanding of the accuracy of the 5*d* state properties in Cs is germane to the ongoing PNC experiment in isoelectronic Ba^+ [15], since the 5*d* state is directly involved in this experiment.

In outline, our approach uses a relativistic all-order method to calculate electric dipole matrix elements for Cs 6*p*-*nd* transitions for $n=5$, 6, and 7. These are used to evaluate 5*d* radiative lifetimes and 6*p* polarizabilities (for the latter, we also include contributions from all other relevant states). Our calculations of the 6*p* scalar polarizabilities, which are in good agreement with experiment, show that they are dominated by contributions from 5*d*-6*p* transitions. These are the only electric dipole transitions contributing to the 5*d* state lifetimes (as we mention below, the 5*d*-6*s* electric quadrupole transition rates are negligibly small). Thus, it is possible to check consistency between polarizability and lifetime measurements by deriving 5*d*-6*p* matrix elements from 5*d* lifetime measurements and substituting these values into the 6*p* polarizability calculations. For either of the two experimental lifetimes, [10,11] this procedure yields a result that disagrees with directly measured polarizabilities [12–14] by several standard deviations.

The particular all-order method used here is the linearized coupled-cluster method which sums infinite sets of many-body perturbation theory terms. We refer the reader to Refs. [16–18] for a detailed description of the approach. The wave function of the valence electron v is represented as an expansion,

$$|\Psi_v\rangle = \left[1 + \sum_{ma} \rho_{ma} a_m^\dagger a_a + \frac{1}{2}\sum_{mnab} \rho_{mnab} a_m^\dagger a_n^\dagger a_b a_a + \sum_{m\neq v} \rho_{mv} a_m^\dagger a_v + \sum_{mna} \rho_{mnva} a_m^\dagger a_n^\dagger a_a a_v + \frac{1}{6}\sum_{mnrab} \rho_{mnrvab} a_m^\dagger a_n^\dagger a_r^\dagger a_b a_a a_v\right]|\Phi_v\rangle, \quad (1)$$

where Φ_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. This lowest-order atomic state function can be written as $|\Phi_v\rangle = a_v^\dagger|0_C\rangle$, where $|0_C\rangle$ represent DHF wave function of a closed core. The indices m, n, and r designate excited states, and indices a and b designate core

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TABLE I. Absolute values of electric dipole $5d$-$6p$ reduced matrix elements in Cs calculated in different approximations: Dirac-Hartree-Fock (DHF), third-order many-body perturbation theory (III), single-double all-order method (SD), single-double all-order method including partial triple contributions (SDpT), and the corresponding scaled values. R is the ratio of the $5d_{3/2}$-$6p_{3/2}$ to $5d_{3/2}$-$6p_{1/2}$ transition matrix elements. All values are given in atomic units (ea_0, where a_0 is the Bohr radius).

Transition	DHF	III	SD	SD_{sc}	SDpT	$SDpT_{sc}$
$5d_{3/2}$-$6p_{1/2}$	8.9784	6.9231	6.5809	7.0634	6.9103	7.0127
$5d_{3/2}$-$6p_{3/2}$	4.0625	3.1191	2.9575	3.1871	3.1112	3.1614
R	0.4525	0.4505	0.4494	0.4512	0.4502	0.4508
$5d_{5/2}$-$6p_{3/2}$	12.1865	9.4545	9.0238	9.6588	9.4541	9.5906

states. The excitation coefficients ρ_{ma}, ρ_{mv}, ρ_{mnab}, and ρ_{mnva} are used to calculate matrix elements, which can be expressed in the framework of the all-order method as linear or quadratic functions of the excitation coefficients. We restrict the expansion given by Eq. (1) to single and double (SD) excitations, with partial inclusion of triple excitations. The results obtained using the SD expansion are referred to as SD data throughout the paper and results obtained with partial addition of the triple excitations are referred to as SDpT data. We also performed third-order many-body perturbation theory calculations, following Ref. [19], to better understand the size of higher-order correlation corrections. Unless stated otherwise, all results in this paper are expressed in the system of atomic units (a.u.).

Table I lists the $5d$-$6p$ reduced electric dipole matrix elements in Cs as calculated using the DHF approximation, third-order perturbation theory (III), single-double all-order method (SD), and single-double all-order method including partial triple contributions (SDpT). We use semi-empirical scaling described, for example, in Ref. [17] to estimate some classes of the omitted high-order corrections. The scaled values are listed in rows labeled SD_{sc} and $SDpT_{sc}$.

We use the $5d$-$6p$ matrix elements from Table I to calculate the lifetimes of the $5d_{3/2}$ and $5d_{5/2}$ levels in Cs. The Einstein A-coefficients A_{vw} are calculated using the formula [19]

$$A_{vw} = \frac{2.02613 \times 10^{15}}{\lambda^3} \frac{|\langle v\|D\|w\rangle|^2}{2j_v+1}\ \mathrm{s}^{-1}, \tag{2}$$

where $\langle v\|D\|w\rangle$ is the reduced electric dipole matrix element for the transition between states v and w and λ is the corresponding wavelength in nanometers. The lifetime of the state v is calculated as

$$\tau_v = \frac{1}{\sum_w A_{vw}}. \tag{3}$$

The results are listed in Table II. The experimental energies from Ref. [22] are used. The scaled SD values are taken as final values based on the comparison of a number of Rb, Cs, and Fr results [23,24] with experiment. The theoretical values differ substantially, by over 5%, from the experimental results (we note that the experimental values from Refs. [10,11] differ by 4%, which exceeds their stated uncertainties of 0.7% and 1%, respectively). One possible source of such a discrepancy is the contribution of the $5d$-$6s$ electric quadrupole transition to the $5d$ lifetime. Our calculation of this rate, using the all-order method, yields a corresponding Einstein A-coefficient for the $5d_{5/2}$-$6s$ transition of 19 Hz, which is only 0.02% of the corresponding electric dipole A-coefficient of 741 kHz (see Table II). Thus, the contribution of the electric quadrupole transition to $5d$ lifetime is entirely negligible within the present experimental and theoretical uncertainties.

To clarify such a large disagreement we check the consistency of the experimental $5d$ lifetime measurements with $6p$ polarizability measurements, which involves contributions from the same transitions. First, we use experimental $5d$ lifetimes from Ref. [10] to determine the $5d$-$6p$ reduced matrix elements. Inverting Eq. (3), we find for the $5d_{5/2}$-$6p_{3/2}$ matrix element, $|\langle 5d_{5/2}\|D\|6p_{3/2}\rangle| = 9.916(35)$. To derive the $5d_{3/2}$-$6p_{1/2}$ and $5d_{3/2}$-$6p_{3/2}$ matrix elements, the lifetime of the $5d_{3/2}$ level alone is not sufficient and some assumption about the ratio R of these matrix elements must be made. We use the theoretical SD_{sc} value 0.4512(18) from Table I for the ratio and assume the deviation of other high-precision theoretical results in Table I from this value to be its uncertainty. The variation of the ratio from one approximation to another is far smaller than the variation in the individual matrix elements, thus the uncertainty is rather low (0.4%). The resulting values of the $5d_{3/2}$-$6p$ matrix elements are the following: $|\langle 5d_{3/2}\|D\|6p_{1/2}\rangle| = 7.283(60)$, $|\langle 5d_{3/2}\|D\|6p_{3/2}\rangle| = 3.286(27)(13)$. We separated the uncertainties in the $5d_{3/2}$-$6p_{3/2}$ matrix elements into contributions from the $5d_{3/2}$ lifetime measurement (0.027) and from the estimation of R (0.013). Combining them, we obtain 3.286(30). The contribution of the uncertainty in R to the uncertainty in the value of $5d_{3/2}$-$6p_{1/2}$ matrix element is negligible.

TABLE II. The values of Einstein A-coefficients A_{vw} (in MHz) and final lifetimes (in ns) for $5d_{5/2}$ and $5d_{3/2}$ states in Cs. The theoretical values are compared with experimental results from Refs. [10,11].

Level	Transition		SD	SD_{sc}	SDpT	$SDpT_{sc}$	Expt. [10]	Expt. [11]
$5d_{5/2}$	$5d_{5/2}$-$6p_{3/2}$	A_{vw}	0.646	0.741	0.710	0.730		
		τ	1547	1350	1409	1369	1281(9)	1226(12)
$5d_{3/2}$	$5d_{3/2}$-$6p_{1/2}$	A_{vw}	0.804	0.926	0.886	0.913		
	$5d_{3/2}$-$6p_{3/2}$	A_{vw}	0.094	0.109	0.104	0.107		
		τ	1114	966	1010	981	909(15)	

The scalar α_0 and tensor α_2 polarizabilities of an atomic state v are calculated using formulas

$$\alpha_0^v = \frac{2}{3(2j_v+1)} \sum_n \frac{\langle n\|D\|v\rangle^2}{E_n - E_v}, \tag{4}$$

$$\alpha_2^v = 4\left(\frac{5j_v(2j_v-1)}{6(j_v+1)(2j_v+1)(2j_v+3)}\right)^{1/2} \times \sum_n (-1)^{j_v+j_n+1} \begin{Bmatrix} j_v & 1 & j_n \\ 1 & j_v & 2 \end{Bmatrix} \frac{\langle n\|D\|v\rangle^2}{E_n - E_v}, \tag{5}$$

where D is the dipole operator and formula for α_0 includes only valence part of the polarizability. The main contributions to the polarizability, $\alpha_{\rm main}$, come from transitions between 6*s*, 7*s*, 8*s*, 9*s*, 6*p*, 7*p*, 8*p*, 9*p*, 5*d*, 6*d*, and 7*d* levels; the remainder, $\alpha_{\rm tail}$, is calculated from summing over all other valence-excited states of the system (which is confined in a sphere of radius $75a_0$). The core contribution to the scalar polarizability $\alpha_{\rm core}=15.8a_0^3$ is taken from Ref. [21], where it was calculated in random-phase approximation (RPA). We note that this value includes the contribution from the valence shell and, therefore, must be compensated by the additional term $\alpha_{\rm vc}$, which is equal to the contribution from the valence shell divided by $(2j_v+1)$ with an opposite sign. We find that the $\alpha_{\rm vc}$ term is negligible for *np* states and very small (below 0.2%) for the 6*s* state. We list the contributions to Cs 6*p* scalar polarizabilities in Table III. The corresponding electric dipole matrix elements *d*, their sources, and uncertainties δd are also given. The values for 6*s*-*np* and 7*s*-*np* transitions are taken from Ref. [6], where the "best value" set of these matrix elements was compiled for the calculation of the tensor transition polarizability β. The 6*p*-6*d* and 6*p*-7*d* matrix elements are from the present *ab initio* SDpT calculation. The values of the 5*d*-6*p* matrix elements are derived from the 5*d* lifetime experiment [10]. The uncertainties of all contributions are listed separately. The uncertainties listed in Ref. [6] are used for 6*s*-*np* and 7*s*-*np* transitions; the difference between SD and SDpT data is taken to be the uncertainty of the 6*p*-6*d* and 6*p*-7*d* matrix elements calculated in this work. The uncertainties of the 5*d*-6*p* matrix elements obtained from the lifetime experiment [10] are derived above. The uncertainty of the core term $\alpha_{\rm core}$ is taken to be 2% based on the comparison of RPA data for closed core systems with experiments and high-precision calculations. The uncertainty of the remaining contribution $\alpha_{\rm tail}$ is estimated to be 30% based on the comparison of the DHF results with correlated values.

We also calculate the scalar polarizability of the 6*s* state using the same methods and data set as for the 6*p* polarizability. The resulting value $\alpha_0(6s)=398.2(0.9)a_0^3$ and its uncertainty are dominated by contributions of the 6*s*-6*p* matrix element taken from experiment of Ref. [20]. We use this result when calculating differences of 6*p* and 6*s* polarizabilities. The recent measurement of the ground state polarizability in Cs yielded the value $\alpha_0(6s)=401.0(0.6)a_0^3$ [25].

We compare the final results for the differences of the 6*p* and 6*s* scalar polarizabilities α_0 and the tensor polarizability α_2 with experiment in Table IV. The results of the above calculation (data from Table III), where we used 5*d*-6*p* matrix elements derived from the 5*d* lifetime experiment are listed in column (a). We find that the difference of the $6p_{3/2}$ and 6*s* scalar polarizabilities which uses numbers for 5*d*-6*p* matrix elements derived from [10] 5*d* lifetime measurements $\alpha_0(6p_{3/2})-\alpha_0(6s)=1322(18)a_0^3$ is inconsistent with both experimental values $1240.2(24)a_0^3$ [13] and $1264(13)a_0^3$ [12]. The difference with first value, which has the smaller uncertainty, is 4.5σ and the difference with the second value is 2.6σ. The difference of the $6p_{1/2}$ and 6*s* scalar polarizabilities which uses numbers for 5*d*-6*p* matrix elements derived from [10] 5*d* lifetime measurements $\alpha_0(6p_{1/2})-\alpha_0(6s)=1006(24)a_0^3$ is also inconsistent with the most recent and most precise experimental value $927.35(12)a_0^3$ [14] by 3.2σ. The value for the $6p_{3/2}$ tensor polarizability $-267.3(4.7)a_0^3$ has much larger uncertainty owing to strong cancellation of the contributions from different transitions, and the difference is 1σ. We note that if we were to use another $5d_{5/2}$

TABLE III. Contributions to the $6p_{1/2}$ and $6p_{3/2}$ scalar polarizabilities α_0 in Cs and their uncertainties $\delta\alpha_0$, in units of a_0^3. The values of corresponding matrix elements *d* (in a.u.), their sources, and uncertainties δd (in %) are also given. The 6*p*-6*d* and 6*p*-7*d* matrix elements are from the present SDpT all-order calculation.

$\alpha_0(6p_{1/2})$	*d*	δd		α_0	$\delta\alpha_0$
$6p_{1/2}$-$5d_{3/2}$	−7.283	0.8	[10][a]	1168.4	18.7
$6p_{1/2}$-6*s*	−4.489	0.1	[20]	−131.9	−0.3
$6p_{1/2}$-$6d_{3/2}$	4.145	4.8	SDpT	110.2	10.6
$6p_{1/2}$-7*s*	−4.236	0.5	[6]	178.4	1.8
$6p_{1/2}$-$7d_{3/2}$	2.033	1.7	SDpT	20.3	0.7
$6p_{1/2}$-8*s*	−1.026	0.6	[6]	5.9	0.1
$6p_{1/2}$-9*s*	0.550	0.5	[6]	1.4	0.0
$\alpha_{\rm tail}$			DHF	35.4	10.6
$\alpha_{\rm core}$			[21]	15.8	0.3
Total				1404	24
$\alpha_0(6p_{3/2})$	*d*	δd		α_0	$\delta\alpha$
$6p_{3/2}$-$5d_{3/2}$	3.286	0.9	[10][a]	142.7	2.6
$6p_{3/2}$-$5d_{5/2}$	9.916	0.3	[10]	1255.5	8.8
$6p_{3/2}$-6*s*	−6.324	0.1	[20]	−124.7	−0.2
$6p_{3/2}$-$6d_{3/2}$	−2.053	4.6	SDpT	14.2	1.3
$6p_{3/2}$-$6d_{5/2}$	−6.010	4.3	SDpT	121.2	10.4
$6p_{3/2}$-7*s*	−6.473	0.5	[6]	225.3	2.3
$6p_{3/2}$-$7d_{3/2}$	−0.969	1.5	SDpT	2.4	0.1
$6p_{3/2}$-$7d_{5/2}$	−2.868	1.4	SDpT	21.0	0.6
$6p_{3/2}$-8*s*	−1.462	0.6	[6]	6.2	0.1
$6p_{3/2}$-9*s*	0.774	0.6	[6]	1.4	0.0
$\alpha_{\rm tail}$			DHF	38.7	11.6
$\alpha_{\rm core}$			[21]	15.8	0.3
Total				1720	18

[a]Derived from the experimental $5d_{3/2}$ lifetime [10] using theoretical ratio of the $6p_{3/2}$-$5d_{3/2}$ and $6p_{1/2}$-$5d_{3/2}$ matrix elements.

TABLE IV. Calculated and experimental values of Cs polarizabilities, in a_0^3. Calculation (a) uses $5d$-$6p$ matrix elements data derived from the $5d$ lifetime experiment [10] (results of Table III); calculation (b) used $5d$-$6p$ theoretical all-order values (SD scaled data). All other contributions in calculations (a) and (b) are the same.

	Present		Expt. [12]	Expt. [13]	Expt. [14]
	(a) Expt. $5d$-$6p$	(b) Theory $5d$-$6p$			
$\alpha_0(6p_{3/2})-\alpha_0(6s)$	1322(18)	1248	1264(13)	1240.2(24)	
$\alpha_0(6p_{1/2})-\alpha_0(6s)$	1006(24)	936	970(9)		927.35(12)
$\alpha_2(6p_{3/2})$	−267(4.7)	−261.2	−261(8)	−262.4(15)	

lifetime experiment [11], the discrepancies with polarizability measurements only increase. Thus, neither $5d_{5/2}$ lifetime experiment [10,11] is consistent with either Refs. [12,13], or [14] Stark shift measurements within the quoted uncertainties.

We calculate that the experimental value of $\alpha_0(6p_{1/2})-\alpha_0(6s)=927.35(12)a_0^3$ [14] corresponds to the lifetime of the $5d_{3/2}$ state $\tau_{5d_{3/2}}=975(14)$ ns, and the experimental value of $\alpha_0(6p_{3/2})-\alpha_0(6s)=1240.2(2.4)a_0^3$ [13] corresponds to the lifetime of the $5d_{5/2}$ state $\tau_{5d_{5/2}}=1359(18)$ ns. The uncertainties in these lifetime values are dominated by the uncertainties in the values of $6p$-$6d$ transitions and the uncertainty in the contribution α_{tail} as evident from Table III.

Finally, we repeated the polarizability calculation by replacing the $5d$-$6p$ matrix elements derived from the lifetime experiment by our theoretical values (SD_{sc}) from Table I. All other matrix elements and contributions are exactly the same as in the first calculation. The results are listed in column (b) of Table IV. As expected, they are quite different from the previous calculation (a) as our theoretical $5d$-$6p$ matrix elements are substantially different from the values derived from $5d$ lifetimes. We find that our theoretical polarizability data are in good agreement (0.4–1%) with experimental values and with the semiempirical results from Ref. [26].

In conclusion, we find the experimental measurements of $5d$ lifetime and $6p$ scalar polarizabilities to be inconsistent within the uncertainties quoted by the experimental groups. Our theoretical calculations are consistent with polarizability experiments but not with the lifetime measurements. Thus, further measurements of the properties of $5d$ and $6p$ states are of great interest for clarification of this issue and for providing benchmark values for $5d$-$6p$ matrix elements.

[1] C. S. Wood *et al.*, Science **275**, 1759 (1997).
[2] S. C. Bennett and C. E. Wieman, Phys. Rev. Lett. **82**, 2484 (1999).
[3] A. Derevianko, Phys. Rev. Lett. **85**, 1618 (2000).
[4] V. A. Dzuba *et al.*, Phys. Rev. A **63**, 044103 (2001).
[5] W. R. Johnson *et al.*, Phys. Rev. Lett. **87**, 233001 (2001).
[6] A. A. Vasilyev *et al.*, Phys. Rev. A **66**, 020101 (2002).
[7] V. A. Dzuba *et al.*, Phys. Rev. D **66**, 076013 (2002).
[8] A. I. Milstein *et al.*, Phys. Rev. Lett. **89**, 283003 (2003).
[9] M. Y. Kuchiev and V. V. Flambaum, J. Phys. B **36**, R191 (2003).
[10] D. DiBerardino *et al.*, Phys. Rev. A **57**, 4204 (1998).
[11] B. Hoeling *et al.*, Opt. Lett. **21**, 74 (1996).
[12] L. R. Hunter *et al.*, Phys. Rev. A **37**, 3283 (1988).
[13] C. E. Tanner and C. Wieman, Phys. Rev. A **38**, 162 (1988).
[14] L. R. Hunter *et al.*, Opt. Commun. **94**, 210 (1992).
[15] T. W. Koerber *et al.*, J. Phys. B **36**, 637 (2003).
[16] S. A. Blundell *et al.*, Phys. Rev. A **40**, 2233 (1989).
[17] S. A. Blundell *et al.*, Phys. Rev. A **43**, 3407 (1991).
[18] M. S. Safronova *et al.*, Phys. Rev. A **60**, 4476 (1999).
[19] W. R. Johnson *et al.*, At. Data Nucl. Data Tables **64**, 279 (1996).
[20] R. J. Rafac *et al.*, Phys. Rev. A **60**, 3648 (1999).
[21] W. R. Johnson *et al.*, At. Data Nucl. Data Tables **28**, 333 (1983).
[22] C. E. Moore, *Atomic Energy Levels*, Natl. Bur. Stand. Ref. Data Ser. Natl. Bur. Stand. (U.S.) Circ. No. 467 (U.S. GPO, Washington, D.C., 1971), Vol. 35.
[23] M. S. Safronova, Ph.D. thesis, University of Notre Dame, 2000.
[24] M. S. Safronova *et al.*, Phys. Rev. A **69**, 022509 (2004).
[25] J. M. Amini and H. Gould, Phys. Rev. Lett. **91**, 153001 (2003).
[26] H. L. Zhou and D. W. Norcross, Phys. Rev. A **40**, 5048 (1989).