

Conforming the measured lifetimes of the $5d^2D_{3/2,5/2}$ states in Cs with theory

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We find very good agreement between our theoretically evaluated lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs with the experimental values reported by DiBerardino *et al.* [*Phys. Rev. A* **57**, 4204 (1998)], which were demonstrated to disagree with an earlier rigorous theoretical study [Safronova and Clark, *Phys. Rev. A* **69**, 040501(R) (2004)] and with the other available precise measurement [Hoeling *et al.*, *Opt. Lett.* **21**, 74 (1996)]. In this work, we carry out calculations of the radiative transition matrix elements using many variants of relativistic many-body methods, mainly in the coupled-cluster theory framework, and analyze the propagation of electron correlation effects to elucidate their roles in accurate evaluations of the matrix elements. We also demonstrate contributions explicitly from Dirac-Coulomb interactions, frequency-independent Breit interaction, and lower order quantum electrodynamics effects. Uncertainties in these matrix elements due to different possible sources of errors are estimated. By combining our calculated radiative matrix elements with the experimental values of the transition wavelengths, we obtain the transition probabilities due to both the allowed and the lower order forbidden channels. Adding these quantities together, the lifetimes of the above two states are determined precisely and plausible reasons for the reported inconsistencies between the earlier theoretical calculations and the experimental results are pointed out.

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I. INTRODUCTION

Alkali-metal atoms in general and Cs atom in particular are suitable for many important experimental studies because of the structure of their energy levels. Many sophisticated measurements can be performed on them using optical magnetometry techniques [1,2]. These atoms couple extremely weakly to the environment, allowing potentially long coherence times. Some of these experiments involve quantum computing [3,4], observation of parity nonconservation (PNC) effects to probe new physics beyond the standard model [5,6], and exotic properties like the nuclear anapole moment [7,8] and measurement of the electric dipole moment due to parity and time reversal symmetry violations [9,10]. In many of these studies, the role of the many-body calculations of various atomic properties is crucial [8,10–15]. In fact, one of the reasons why a number of experiments have been carried out on alkali-metal atoms is that they can test the capabilities of the many-body methods employed to evaluate the properties of the ground and excited states of these atoms to a very high precision. This bolsters confidence in using the results obtained from these methods to gain insights into many fundamental physics phenomena [7,8,10–12]. Earlier results from some of the methods have been used for this purpose, but in a few cases, there have been large disagreements between the theoretical results obtained using these methods and their corresponding experimental values: especially when studying properties of the excited states. This provides the opportunity to investigate the missing underlying physics that may shed some light on and may open up windows to probing new physics. One such example is the disagreement between the calculations and the experimental results for the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs [13]. At least two precise

measurements have been carried out to determine the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs [16,17], among which the lifetime of the $5d^2D_{5/2}$ state has been reported as 1281(9) ns [16] and 1225(12) ns [17], values which are clearly out of the ranges of their reported error bars. On the other hand, the difficulty in performing precise calculations of the excited-state properties lies in the important role played by the continuum states in the evaluation of the atomic wave function. Moreover, determination of the wave functions of excited states with a large orbital angular momentum (l), e.g., D states, requires a large number of determinantal states for which an approach like the configuration interaction (CI) method approximated only at singles and doubles excitations (CISD method) may not be suitable for estimating their properties very accurately [18,19]. However, the relativistic coupled-cluster (RCC) method even at the same level of singles and doubles approximation (CCSD method) is capable of estimating many atomic properties within a reasonable accuracy [20–22]. One of the unique features of the (R)CC method is that it can capture the higher excited determinantal states just by approximating the method with only the singles and doubles excitations, owing to the exponential structure of the wave function [22–25]. For example, the CCSD method still accounts for contributions from many of the triples, quadruples, etc., which can be represented by determinantal states containing nonlinear terms [24,25]. To achieve high-accuracy results for excited states with large l values, inclusion of the contributions from these triple and quadruple determinantal states is imperative. However, the CCSD method with only linear terms (LCCSD method) supplemented with other corrections has often been employed in calculations of the atomic properties of alkali-metal atoms to avoid the requirement of large computational resources to account for the nonlinear RCC terms (e.g., see Ref. [13] and references therein). Thus, comparative studies of the theoretical and experimental results of the excited-state atomic properties of alkali-metal atoms using a many-body method

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would serve as a good test of its capability to describe the results accurately.

This work is intended to probe again the validity of the reported experimental values of the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs which, as mentioned above, are inconsistently reported in different works [13]. Experimental values from Refs. [16,17] are discussed in Ref. [13], and a few more measured values with large error bars are also available in the literature [26–28]. Similarly, a few more calculations of the lifetimes of the above states are also reported in the literature [29–33], among which many of them are estimated using the nonrelativistic theory and semiempirical approaches. To appreciate the consideration of extra physical effects in the present work and to realize the reason for observing inconsistencies among the earlier theoretical and experimental results, here we briefly discuss the approximation in the method undertaken in Ref. [13]. In Ref. [13], the SD method, which is equivalent to the LCCSD method, and the SD method with important partial triples corrections (SDpT method) were employed to calculate the electric dipole ($E1$) matrix elements. These values were further revamped by scaling the wave functions of the SD method (SD_{sc} method) and of the SDpT method (SDpT_{sc} method). Then the scalar dipole polarizabilities of the $6p^2P_{1/2}$ and $6p^2P_{3/2}$ states of Cs were evaluated in two ways: first, using the $E1$ matrix elements only from the SDpT method and, second, combining the $E1$ matrix elements extracted from the measured lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states from Refs. [16] with a few other $E1$ matrix elements from the SDpT method. The differential polarizabilities of these $6P$ states with respect to the ground state were determined by taking the precisely measured ground-state dipole polarizability as $401.0(6) ea_0^3$ [34], for the Bohr radius a_0 . These differential polarizabilities were then compared with the available direct measurements [35–37]. This comparison demonstrated that the values obtained using the $E1$ matrix elements entirely from the SDpT method match better with the experimental results than the values obtained using the combination approach. On this basis it was argued that the measured values of the lifetimes of the $5D$ states and the differential polarizabilities of the $6P$ states were inconsistent. For this reason it was assumed that the $E1$ matrix elements obtained using the SDpT method are more accurate and the $E1$ matrix elements deduced from the measured lifetimes were inappropriate. This indirectly meant that the measured lifetimes are invalid. Using the $E1$ matrix elements from the SD method, the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states were obtained as 1114 and 1547 ns, respectively [13]. These values were changed to 966 and 1350 ns for the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states, respectively, when the matrix elements were improved using the SD_{sc} method. Similarly, the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states were obtained as 1010 and 1409 ns, respectively, upon the use of the matrix elements from the SDpT method. Finally, they were obtained as 981 and 1369 ns, respectively, using the matrix elements from the SDpT_{sc} method.

In the above analysis, two things need to be carefully scrutinized further. First, it should be noted that after scaling the wave functions the results were changed significantly. It is very difficult to justify the accuracies of the results from the first principle by scaling the wave functions. Large

differences between the $E1$ matrix elements obtained before and those obtained after scaling the wave functions indicate that it is necessary to consider more physical effects in the above employed SD or SDpT method to improve the accuracies of the results. Second, it can be found, using the sum-over-states approach employed in Ref. [13] to evaluate the polarizabilities of the $6P$ states, that the principal contributions come from the $E1$ matrix elements among the $6p^2P_{1/2} \rightarrow 5d^2D_{3/2}$, $6p^2P_{3/2} \rightarrow 5d^2D_{3/2}$, and $6p^2P_{3/2} \rightarrow 5d^2D_{5/2}$ transitions. However, ample contributions also come from the $E1$ matrix elements involving other D excited states. Again, the sum-over-states approach has limitations accounting for contributions from the core orbitals, higher excited states, and continuum accurately. In order to verify the accuracies of the theoretically estimated lifetimes of the $5D$ states, we would like to carry out calculations of the $E1$ matrix elements of the $6p^2P_{1/2} \rightarrow 5d^2D_{3/2}$, $6p^2P_{3/2} \rightarrow 5d^2D_{3/2}$, and $6p^2P_{3/2} \rightarrow 5d^2D_{5/2}$ transitions considering more physical effects than the LCCSD method: especially through the nonlinear terms of the CCSD method. However, we also employ other lower order many-body methods such as the Dirac-Hartree-Fock (DHF) method, lower order perturbation theory (MBPT method), and LCCSD method to demonstrate gradual changes in the results with the propagation of correlation effects through the higher order terms. We also give the contributions from the important triple excitations in a variety of procedures and, from a semiempirical approach, by using the experimental energies in the calculations of the wave functions in the RCC method. Again, the Dirac-Coulomb (DC) Hamiltonian was considered in Ref. [13]. We also estimate corrections due to the higher order relativistic effects by considering the Breit interaction and lower order quantum electrodynamics (QED) effects in the calculations.

Many general implications for studying the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states in Cs have been discussed previously. We, however, place emphasis on only two applications here, for which the present work could be directly relevant. The most precise PNC measurement has been carried out in the $6s^2S_{1/2} \rightarrow 7s^2S_{1/2}$ transition of Cs, but the nuclear parameters inferred from the nuclear anapole moment deduced from this measurement disagree with the values given by the well-established nuclear models [7,8]. This urges further investigation of PNC effects in atomic systems. In fact, theoretical study demonstrates that the PNC amplitude in the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition is almost three times higher than the $6s^2S_{1/2} \rightarrow 7s^2S_{1/2}$ transition in Cs [14,15]. A plausible principle for measuring the PNC-induced frequency shift in the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition of Ba⁺ has been described in [38]. Following this, it has also been suggested that the same principle can be adopted to measure the PNC effect in the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2}$ transition of Cs [39]. In another work, it has been highlighted that the measurement of PNC-induced light shifts in the S - $D_{5/2}$ transitions of atomic systems would provide an unambiguous signature of the existence of the nuclear anapole moment [40,41]. One of the requirements for enhancing the signal-to-noise ratio in the PNC-induced light-shift measurement principle is to have longer lifetimes of the states involved in the transition [38]. Thus, it is necessary to ensure reliability of the observed lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states in the case where the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2,5/2}$

transitions in Cs are undertaken for PNC measurements. In fact, it has also been advocated that the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs are very suitable for the resonance ionization spectroscopy process owing to their longer lifetimes [42]. Therefore, it is important that ambiguity in the correctness of the lifetime values of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs is eliminated to establish their real values.

II. THEORY

In the Cs atom, it is obvious to assume that the dominant emission transition probabilities for an electron to jump from the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states are due to the $E1$ channel to the low-lying $6p^2P_{1/2}$ and $6p^2P_{3/2}$ states. Since the aim of the present work is to explain the cause of disagreement between the previous theoretical calculations and the experimental results, we intend to show how low the transition probabilities really are due to the forbidden channels from the above two states. Thus, we also determine the transition probabilities due to the next dominant magnetic dipole ($M1$) and electric quadrupole ($E2$) channels from the $5d^2D_{3/2}$ state to the ground state $6s^2S_{1/2}$ and from the $5d^2D_{5/2}$ state to the ground and $5d^2D_{3/2}$ states. The general expressions for evaluating these transition probabilities between the $|\Psi_i\rangle \rightarrow |\Psi_f\rangle$ transitions are given by

$$A_{if}^{E1} = \frac{2.0261 \times 10^{-6}}{\lambda_{if}^3 g_i} S_{if}^{E1}, \quad (1)$$

$$A_{if}^{M1} = \frac{2.6971 \times 10^{-11}}{\lambda_{if}^3 g_i} S_{if}^{M1}, \quad (2)$$

and

$$A_{if}^{E2} = \frac{1.1195 \times 10^{-22}}{\lambda_{if}^5 g_i} S_{if}^{E2}, \quad (3)$$

where the quantity $S_{if}^O = |\langle \Psi_i | O | \Psi_f \rangle|^2$ is known as the line strength for the corresponding reduced matrix element $|\langle \Psi_i | O | \Psi_f \rangle|$ of a transition operator O . These quantities are given later in the paper, in atomic units (a.u.). In the above expressions, $g_i = 2J_i + 1$ is the degeneracy factor of state $|\Psi_i\rangle$ with the angular momentum of state J_i and the transition wavelength (λ_{if}) is used, in nanometers (nm), and when substituted, the transition probabilities (A_{if}^O s) are obtained in units of the inverse of a second (s^{-1}). The lifetime (τ_i) of atomic state $|\Psi_i\rangle$ is determined (in s) by taking the reciprocal of the total emission transition probabilities due to all possible channels, i.e.,

$$\tau_i = \frac{1}{\sum_{O,f} A_{if}^O}, \quad (4)$$

where the summations over O and f correspond to all the decay channels and all the lower states, respectively.

The reduced matrix elements for the $E1$, $M1$, and $E2$ transition operators in terms of the single-particle orbitals are given by

$$\begin{aligned} \langle \kappa_f || e1 || \kappa_i \rangle &= \frac{3}{k} \langle \kappa_f || C^{(1)} || \kappa_i \rangle \int_0^\infty dr \left(j_1(kr) \right. \\ &\quad \left. \times [P_f(r)P_i(r) + Q_f(r)Q_i(r)] + j_2(kr) \right. \end{aligned}$$

$$\begin{aligned} &\times \left\{ \frac{\kappa_f - \kappa_i}{2} [P_f(r)Q_i(r) + Q_f(r)P_i(r)] \right. \\ &\quad \left. + [P_f(r)Q_i(r) - Q_f(r)P_i(r)] \right\}, \quad (5) \end{aligned}$$

$$\begin{aligned} \langle \kappa_f || m1 || \kappa_i \rangle &= \frac{6}{\alpha k} \frac{(\kappa_f + \kappa_i)}{2} \langle -\kappa_f || C^{(1)} || \kappa_i \rangle \int_0^\infty dr \\ &\quad \times j_1(kr) [P_f(r)Q_i(r) + Q_f(r)P_i(r)], \quad (6) \end{aligned}$$

and

$$\begin{aligned} \langle \kappa_f || e2 || \kappa_i \rangle &= \frac{15}{k^2} \langle \kappa_f || C^{(2)} || \kappa_i \rangle \int_0^\infty dr \left(j_2(kr) \right. \\ &\quad \times [P_f(r)P_i(r) + Q_f(r)Q_i(r)] + j_3(kr) \\ &\quad \times \left\{ \frac{\kappa_f - \kappa_i}{3} [P_f(r)Q_i(r) + Q_f(r)P_i(r)] \right. \\ &\quad \left. + [P_f(r)Q_i(r) - Q_f(r)P_i(r)] \right\}, \quad (7) \end{aligned}$$

where $P(r)$ and $Q(r)$ denote the large and small components of the radial parts of the single-particle Dirac orbitals, respectively, the κ s are their relativistic angular momentum quantum numbers, α is the fine-structure constant $k = \alpha(\epsilon_f - \epsilon_i)$ with the orbital energies ϵ s, and $j_l(kr)$ is the spherical Bessel function. The reduced Racah coefficients of rank k are given by

$$\begin{aligned} \langle \kappa_f || C^{(k)} || \kappa_i \rangle &= (-1)^{j_f+1/2} \sqrt{(2j_f+1)(2j_i+1)} \\ &\quad \times \begin{pmatrix} j_f & k & j_i \\ 1/2 & 0 & -1/2 \end{pmatrix} \pi(l_{\kappa_f}, k, l_{\kappa_i}), \quad (8) \end{aligned}$$

with

$$\pi(l, k, l') = \begin{cases} 1 & \text{for } l+k+l' = \text{even,} \\ 0 & \text{otherwise} \end{cases} \quad (9)$$

for the orbital momentum l_κ of the corresponding orbital having the relativistic quantum number κ .

For the first time, we use a different type of analytical basis function having the quadratic type of exponents to express the single-particle wave functions (defined as quadratic-type orbitals) to calculate the above reduced matrix elements. Using these functions, the radial components of the orbitals are expressed as

$$P(r) = \sum_{v=1}^{N_l} c_v^P \mathcal{N}_v^P r^l e^{-\eta_v r^4}$$

and

$$Q(r) = \sum_{v=1}^{N_l} c_v^Q \mathcal{N}_v^Q r^l \left(\frac{d}{dr} + \frac{\kappa}{r} \right) [r^l e^{-\eta_v r^4}], \quad (10)$$

where N_l represents the total number of quadratic-type orbitals considered in the calculations, η_v is an arbitrary coefficient suitably chosen to obtain wave functions accurately, $c_v^{P(Q)}$ values are the linear combination coefficients, and $\mathcal{N}_v^{P(Q)}$ is the normalization constant of the v th basis function for the large (small) component of the wave function. It can be noted that the kinetic balance condition between the large and the small components has been implemented above. The normalization

TABLE I. List of the parameters considered to define the basis functions using quadratic-type orbitals in the present calculations.

	s	p	d	f	g
N_l	34	33	32	31	30
η_0	2.0×10^{-8}	2.5×10^{-8}	2.5×10^{-8}	2.1×10^{-1}	2.1×10^{-7}
ζ	4.67	4.78	4.93	7.08	8.25

constants are given by

$$\mathcal{N}_v^P = 2(2\eta_v)^{\frac{2l+1}{8}} \Gamma\left(\frac{2l+1}{4}\right)^{-1/2} \quad (11)$$

and

$$\mathcal{N}_v^Q = \left[\frac{(l+\kappa)^2}{4(2\eta_v)^{\frac{2l-1}{4}}} \Gamma\left(\frac{2l-1}{4}\right) - \frac{2(l+\kappa)}{(2\eta_v)^{\frac{2l-1}{4}}} \right. \\ \left. \times \Gamma\left(\frac{2l+3}{4}\right) + \frac{4}{(2\eta_v)^{\frac{2l-1}{4}}} \Gamma\left(\frac{2l+7}{4}\right) \right]^{-1}. \quad (12)$$

For convenience, the η_v parameters are constructed to satisfy the even tempering condition between the two parameters η_0 and ζ as

$$\eta_v = \eta_0 \zeta^{v-1}. \quad (13)$$

In Table I we list the η_0 and ζ parameters used in the present calculations.

III. MANY-BODY METHODS

In our previous work [20], we have described the general procedures of our MBPT(2) and RCC methods from which we have calculated the wave functions and transition matrix elements of the Fr atom in the approach of Bloch's formalism [23]. We also adopt these methods here, along with a few more variants of the RCC methods, by approximating the levels of excitations and nonlinear terms in the expression of the wave function. We apply these methods in order to investigate the reason for the discrepancies between the previous theoretical study and the experimental results [13]. We discuss these methods briefly below to illustrate distinctly the roles of the higher order correlation effects in enhancing the accuracies of the calculations of the transition matrix elements.

In Bloch's prescription the atomic wave function of a state $|\Psi_v\rangle$ of Cs with valence orbital v is expressed as [23]

$$|\Psi_v\rangle = \Omega_v |\Phi_v\rangle, \quad (14)$$

where Ω_v and $|\Phi_v\rangle$ are referred to as the wave operator and the reference state, respectively. For computational simplicity we choose the working reference state as the DHF wave function $|\Phi_c\rangle$ for the closed-shell configuration $[5p^6]$, which is common to the ground and the excited states that are involved in the estimations of the lifetimes of the $5D$ states of Cs. Then the actual reference state is constructed from it as $|\Phi_v\rangle = a_v^\dagger |\Phi_c\rangle$ for the respective state with valence orbital v . First, the calculations are performed using the DC Hamiltonian, which

(in a.u.) is given by

$$H = \sum_i \left[c\alpha_i \cdot \mathbf{p}_i + (\beta_i - 1)c^2 + V_n(r_i) + \sum_{j>i} \frac{1}{r_{ij}} \right], \quad (15)$$

where α and β are the usual Dirac matrices and $V_n(r)$ represents the nuclear potential. We evaluate the nuclear potential considering the Fermi-charge distribution defined by

$$\rho_n(r) = \frac{\rho_0}{1 + e^{(r-b)/a}} \quad (16)$$

for the normalization factor ρ_0 , the half-charge radius b , and $a = 2.3/4(\ln 3)$ is related to the skin thickness. We have used $a = 2.3/4(\ln 3)$ and $b = 5.6707$ fm, which is determined using the relation

$$b = \sqrt{\frac{5}{3}r_{\text{rms}}^2 - \frac{7}{3}a^2\pi^2}, \quad (17)$$

with the root mean square (rms) charge radius of the nucleus determined using the formula

$$r_{\text{rms}} = 0.836A^{1/3} + 0.570 \quad (18)$$

(in fm) for the atomic mass A .

Contributions from the frequency-independent Breit interaction are estimated by adding the corresponding interaction term, given by

$$V_B(r_{ij}) = -\frac{1}{2r_{ij}} \{\alpha_i \cdot \alpha_j + (\alpha_i \cdot \hat{\mathbf{r}}_{ij})(\alpha_j \cdot \hat{\mathbf{r}}_{ij})\}. \quad (19)$$

We have also estimated corrections from the lower order QED effects by considering the following potentials with H in a formalism similar to that described in Ref. [43], but for the nuclear Fermi-charge distribution given above. The lower order vacuum polarization (VP) effects are considered at the approximations of the Uehling $[V_U(r)]$ and Wichmann-Kroll $[V_{\text{WK}}(r)]$ potentials, which are given by

$$V_U(r) = -\frac{2\alpha^2}{3r} \int_0^\infty dx x \rho_n(x) \int_1^\infty dt \sqrt{t^2 - 1} \\ \times \left(\frac{1}{t^3} + \frac{1}{2t^5} \right) [e^{-2ct|r-x|} - e^{-2c(r+x)}] \quad (20)$$

and

$$V_{\text{WK}}(r) = -\frac{8Z^2\alpha^4}{9r} 0.092 \int_0^\infty dx x \rho_n(x) \\ \times (0.22\{\arctan[1.15(-0.87 + 2c|r-x|)] \\ - \arctan[1.15(-0.87 + 2c(r+x))]\} \\ + 0.22\{\arctan[1.15(0.87 + 2c|r-x|)] \\ - \arctan[1.15(0.87 + 2c(r+x))]\} \\ - 0.11\{\ln[0.38 - 0.87c|r-x| + c^2(r-x)^2] \\ - \ln[0.38 - 0.87c(r+x) + c^2(r+x)^2]\} \\ + 0.11\{\ln[0.38 + 0.87c|r-x| + c^2(r-x)^2] \\ - \ln[0.38 + 0.87c(r+x) + c^2(r+x)^2]\}), \quad (21)$$

with the atomic number of the system Z . The contribution from the self-energy (SE) interaction is accounted for by evaluating

contributions together from the electric form factor, given by

$$V_{SE}^{ef}(r) = -A(Z)(Z\alpha)^4 e^{-Zr} + \frac{B(Z,r)\alpha^2}{r} \int_0^\infty dx x \rho_n(x) \\ \times \int_1^\infty dt \frac{1}{\sqrt{t^2-1}} \left\{ \left(\frac{1}{t} - \frac{1}{2t^3} \right) \right. \\ \times \left[\ln(t^2-1) + 4 \ln \left(\frac{1}{Z\alpha} + \frac{1}{2} \right) \right] - \frac{3}{2} + \frac{1}{t^2} \left. \right\} \\ \times [e^{-2ct|r-x|} - e^{-2ct(r+x)}], \quad (22)$$

and from the magnetic form factor, given by

$$V_{SE}^{mg}(r) = \frac{i\alpha}{4\pi c} \boldsymbol{\gamma} \cdot \nabla_r \int_0^\infty d^3x \rho_n(x) \\ \times \left\{ \left(\int_1^\infty dt \frac{e^{-2trR}}{Rt^2\sqrt{t^2-1}} \right) - \frac{1}{R} \right\}, \quad (23)$$

where $A(Z) = 0.074 + 0.35Z\alpha$, $B(Z,r) = \{1.071 - 1.97[(Z-80)\alpha]^2 - 2.128[(Z-80)\alpha]^3 + 0.169[(Z-80)\alpha]^4\}cr/[cr + 0.07(Z\alpha)^2]$, and $R = |\mathbf{r} - \mathbf{x}|$.

Following the form of the reference states in our approach, Ω_v can now be divided as

$$\Omega_v = 1 + \chi_c + \chi_v, \quad (24)$$

where χ_c and χ_v are responsible for carrying out the excitations from $|\Phi_c\rangle$ and $|\Phi_v\rangle$, respectively, due to the residual interaction $V_r = H - H_0$ for the DHF Hamiltonian H_0 . In a perturbative series expansion, we can express

$$\chi_c = \sum_k \chi_c^{(k)} \quad \text{and} \quad \chi_v = \sum_k \chi_v^{(k)}, \quad (25)$$

where the superscript k refers to the number of times V_r is considered in the MBPT method [denoted the MBPT(k) method]. The k th-order amplitudes for the χ_c and χ_v operators are obtained by solving the equations [23]

$$[\chi_c^{(k)}, H_0]P = QV_r(1 + \chi_c^{(k-1)})P \quad (26)$$

and

$$[\chi_v^{(k)}, H_0]P = QV_r(1 + \chi_c^{(k-1)} + \chi_v^{(k-1)})P - \sum_{m=1}^{k-1} \chi_v^{(k-m)} \\ \times PV_r(1 + \chi_c^{(m-1)} + \chi_v^{(m-1)})P, \quad (27)$$

where the projection operators $P = |\Phi_c\rangle\langle\Phi_c|$ and $Q = 1 - P$ describe the model space and the orthogonal space of the DHF Hamiltonian H_0 , respectively. The energy of state $|\Psi_n\rangle$ is evaluated using an effective Hamiltonian,

$$H_v^{\text{eff}} = Pa_v H \Omega_v a_v^\dagger P. \quad (28)$$

Using the normal-order Hamiltonian $H_N = H - PHP$ in place of H in the above expression, the attachment energy of a state with valence orbital v is evaluated.

In the (R)CC theory ansatz, the wave functions of the considered states are expressed as

$$|\Psi_v\rangle \equiv \Omega_v |\Phi_v\rangle = e^T \{1 + S_v\} |\Phi_v\rangle, \quad (29)$$

with $\chi_c = e^T - 1$ and $\chi_v = e^T S_v - 1$, where T and S_v are the CC excitation operators that excite electrons from the core and core along with the valence orbitals in the virtual space,

respectively. In this work, we have considered only single and double excitations, denoted by the subscripts 1 and 2, respectively, in the CCSD method by expressing

$$T = T_1 + T_2 \quad \text{and} \quad S_v = S_{1v} + S_{2v}. \quad (30)$$

In the LCCSD method, only the linear terms are retained as in the SD method in Ref. [13]. The amplitudes of these operators are evaluated using the equations

$$\langle\Phi_c^* | \bar{H}_N | \Phi_c\rangle = 0 \quad (31)$$

and

$$\langle\Phi_v^* | (\bar{H}_N - \Delta E_v) S_v | \Phi_v\rangle = -\langle\Phi_v^* | \bar{H}_N | \Phi_v\rangle, \quad (32)$$

where $|\Phi_c^*\rangle$ and $|\Phi_v^*\rangle$ are the excited-state configurations, here up to doubles, with respect to DHF states $|\Phi_c\rangle$ and $|\Phi_v\rangle$, respectively, and $\bar{H}_N = (H_N e^T)_l$ with subscript l represents the linked terms only. Here $\Delta E_v = H_v^{\text{eff}} - H_c^{\text{eff}}$ is the attachment energy of the electron in the valence orbital v with $H_c^{\text{eff}} = PH(1 + \chi_c)P$. Following Eq. (28), the expression for ΔE_v is given by

$$\Delta E_v = \langle\Phi_v | \bar{H}_N \{1 + S_v\} | \Phi_v\rangle. \quad (33)$$

We also include contributions from the important triply excited configurations by defining perturbative operators as

$$T_3^{\text{pert}} = \frac{1}{6} \sum_{abc,pqr} \frac{(H_N T_2)_{abc}^{pqr}}{\epsilon_a + \epsilon_b + \epsilon_c - \epsilon_p - \epsilon_q - \epsilon_r} \quad (34)$$

and

$$S_{3v}^{\text{pert}} = \frac{1}{4} \sum_{ab,pqr} \frac{(H_N T_2 + H_N S_{2v})_{abv}^{pqr}}{\epsilon_a + \epsilon_b + \epsilon_v - \epsilon_p - \epsilon_q - \epsilon_r}, \quad (35)$$

where $\{a,b,c\}$ and $\{p,q,r\}$ represent the occupied and virtual orbitals, respectively, and the ϵ s are their corresponding orbital energies. Since the final results reported in Ref. [13] use the SDpT method and scale the wave functions, we would like to determine the roles of the triply excited configurations in the evaluation of transition matrix elements. However, the exact procedure using which triple excitations accounted in the SDpT method is not clear to us, so we try to estimate these contributions in various ways. When the S_{3v}^{pert} operator is considered as a part of the S_v operator to estimate only the energies using Eq. (33) after obtaining the RCC amplitudes, this is referred to as the L/CCSD(T) method. However, when it is required to estimate both the energies and the amplitudes of the S_v operators simultaneously in the iterative procedure through Eqs. (32) and (33), we call this the L/CCSD[T] method. To explore the roles of the core correlations through the triple excitations, we consider the T_3^{pert} operator as a part of the T operator when solving Eq. (31). This is referred to as the L/CCSDpT^c method, and when, along with this approach, the S_{3v}^{pert} operator is considered in Eqs. (32) and (33), we refer to this as the L/CCSDpT method. But we consider both the T_3^{pert} and the S_{3v}^{pert} operators in Eqs. (31) and (32) only to ameliorate the amplitudes of the T_1 and S_{1v} operators for computational ease.

After obtaining the amplitudes of the MBPT and RCC operators using the equations described earlier, the transition

matrix element of an operator O between state $|\Psi_i\rangle$ and state $|\Psi_f\rangle$ is evaluated using the expression

$$\frac{\langle\Psi_f|O|\Psi_i\rangle}{\sqrt{\langle\Psi_f|\Psi_f\rangle\langle\Psi_i|\Psi_i\rangle}} = \frac{\langle\Phi_f|\Omega_f^\dagger O\Omega_i|\Phi_i\rangle}{\sqrt{\langle\Phi_f|\Omega_f^\dagger\Omega_f|\Phi_f\rangle\langle\Phi_i|\Omega_i^\dagger\Omega_i|\Phi_i\rangle}}. \quad (36)$$

This gives rise to a finite number of terms for the MBPT(2) and LCCSD-variant methods, but it involves two nonterminating series in the numerator and denominator, $e^{T^\dagger} O e^T$ and $e^{T^\dagger} e^T$, respectively, in the CCSD-variant methods. As described in our previous works [20,44,45], we adopt iterative procedures to account for the contributions from these nontruncative series. For a comprehensive understanding, we also give intermediate results keeping k number of T and/or T^\dagger operators in these series using the CCSD method to evaluate the matrix elements, and we refer to this approach as the CCSD^(k) method. Finally, our CCSD results correspond to the calculations using the CCSD^(∞) method. We also estimate the contributions due to triple excitations by considering both the T_3^{pert} and the S_{3v}^{pert} operators along with their complex conjugates in Eq. (36) of the L/CCSD method and refer to this approach as the L/CCSD_{t3} method.

IV. RESULTS AND DISCUSSION

Before presenting the transition matrix elements obtained with various methods, we would first like to validate the methods by carrying out calculations of the attachment energies of the considered states of Cs and comparing them against their corresponding experimental values. Although it is understood that the nature of the radial parts of the wave functions could be different for accurate evaluation of the energies and transition matrix elements, it can be noted from Eqs. (27) and (32) that the energy-evaluating expressions are also coupled with the wave-function-determining equations. Hence, accurate evaluation of the energies using a method can be an indication for verifying the validity of the method in addition to accounting for more physical effects in the method. For this purpose, we list the energies obtained with various methods, described previously, in Table II using the DC Hamiltonian and compare them with the experimental values [46]. We find that the CCSD method gives rise to fairly accurate results for all the states considered. It is also noted that the MBPT(2) values are more accurate than the LCCSD values, but the partial triple effects bring the LCCSD results closer to the experimental values. When these partial effect contributions are added to the CCSD results, the results become far off from the experimental results. This, therefore, implies that the neglected triples effects, which can contribute mainly through the T_2 and S_{2v} amplitude-determining equations, may cancel out some of these overestimated triple excited contributions to give, finally, more precise results than accounting for them only through the singles excitations. We also observe that the triples effects through the valence orbital excitations are the dominant ones over the core-triple excitations. Nevertheless, it would be pertinent to consider full triple excitations in this situation rather than adapting them through the partial effects. Therefore, we consider the results from the CCSD

TABLE II. Demonstration of trends of the calculated energies (in cm^{-1}) using various relativistic methods considered in the present work with the DC Hamiltonian. Relativistic corrections are given separately from the CCSD method. These results are compared with the experimental values [46]. Uncertainties in the experimental values are not mentioned, as they are more precise than the quoted values up to the second decimal places. Boldface font is to highlight accuracies in the results obtained from the calculations with respect to the experimental values.

Method	$6s^2S_{1/2}$	$6p^2P_{1/2}$	$6p^2P_{3/2}$	$5d^2D_{3/2}$	$5d^2D_{5/2}$
DHF	27983.73	18752.17	18350.36	14096.82	14121.80
MBPT(2)	32020.63	20362.23	19777.17	16681.89	16568.09
LCCSD	32425.64	20566.54	19965.95	17882.53	17718.94
LCCSD(T)	31812.61	20335.35	19762.15	17439.99	17325.21
LCCSDpT ^c	32425.66	20566.54	19965.95	17882.53	17718.94
LCCSD[T]	31834.43	20340.48	19766.15	17505.35	17381.71
LCCSDpT	31758.84	20310.25	19741.51	17374.41	17281.31
CCSD	31463.22	20159.54	19600.28	16537.86	16445.08
CCSD(T)	31090.05	20011.04	19470.48	16259.53	16149.22
CCSDpT ^c	31428.69	20149.29	19591.82	16504.24	16414.53
CCSD[T]	31064.13	20013.07	19472.02	16272.98	16214.82
CCSDpT	31001.99	19993.26	19455.22	16223.73	16170.23
Relativistic corrections					
Breit	-0.40	-7.50	-1.32	20.17	23.62
VP	3.63	-0.03	-0.09	-0.40	-0.36
SE	-17.92	-1.09	0.95	2.11	2.15
Breit + QED	-14.86	-8.62	-0.47	21.87	25.42
Experiment	31406.47	20228.20	19674.26	16907.21	16809.62

method, which accounts for all the nonlinear terms within the considered level of excitations, as the recommended calculated values for further use.

We have also explicitly estimated the contributions due to the Breit interaction (“Breit”), the VP effect (“VP”), and the SE effect (“SE”) using the CCSD method; they are listed towards the bottom of Table II. In addition, we also determine these corrections considering all these relativistic corrections together with respect to the contributions from the DC Hamiltonian in the CCSD method (“Breit + QED”). We find slight changes in the results obtained with the Breit + QED approach versus those obtained when the corrections estimated independently are added up. We observe that among all these relativistic corrections, the SE effect is large in the ground state while the Breit interaction gives larger corrections in the other considered states.

After analyzing the energies obtained with various methods with respect to the experimental values, in Table III we list the $E1$ matrix elements of the transitions that are required to estimate the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs. We compare these results with the values reported recently by other groups using different relativistic many-body methods [13,14]. In addition to the methods we have employed to evaluate the energies, we also list the $E1$ matrix elements using the L/CCSD_{t3} and L/CCSD_{ex} methods in the Table III. We find reasonable agreement between the results obtained using our DHF and LCCSD methods and those obtained with the DHF and SD methods in Ref. [13]. However, there are significant differences in the results when the higher order effects are

TABLE III. Our $E1$ reduced matrix elements (in a.u.) from various methods and comparison with the other relativistic calculations. Relativistic corrections to our results are quoted separately and the recommended values are also listed.

Method	$5d_{3/2} \rightarrow 6p_{1/2}$	$5d_{3/2} \rightarrow 6p_{3/2}$	$5d_{5/2} \rightarrow 6p_{3/2}$
DHF	9.012	4.078	12.233
MBPT(2)	7.535	3.404	10.273
LCCSD	6.566	2.954	9.011
LCCSD ₁₃	6.569	2.952	9.015
LCCSDpT ^c	6.566	2.954	9.011
LCCSD[T]	6.472	2.909	8.899
LCCSDpT	6.687	3.009	9.137
LCCSD _{ex}	6.305	2.828	8.683
CCSD ⁽²⁾	7.292	3.291	9.931
CCSD ⁽⁴⁾	7.301	3.295	9.941
CCSD^(∞)	7.301	3.295	9.941
CCSD ₁₃	7.304	3.293	9.945
CCSDpT ^c	7.326	3.307	10.018
CCSD[T]	7.258	3.275	9.934
CCSDpT	7.357	3.320	10.056
CCSD _{ex}	7.348	3.318	10.050
Relativistic corrections			
Breit	-0.009	-0.005	0.022
VP	~0.0	~0.0	0.039
SE	-0.001	-0.001	0.037
Breit + QED	-0.010	-0.005	0.020
Estimated uncertainties			
Basis	0.048	0.023	0.022
Triples	0.003	0.002	0.004
Scaling	0.047	0.023	0.109
Recommended values			
Recommended	7.291(67)	3.288(33)	9.961(111)
From Ref. [13]			
DHF	8.9784	4.0625	12.1865
MBPT(3)	6.9231	3.1191	9.4545
SD	6.5809	2.9575	9.0238
SD _{sc}	7.0634	3.1871	9.6588
SDpT	6.9103	3.1112	9.4541
SDpT _{sc}	7.0127	3.1614	9.5906
From Ref. [14]			
$\Sigma^{(2)}$	6.744	3.037	9.254
$\lambda \Sigma^{(2)}$	7.039	3.173	9.629
$\Sigma^{(\infty)}$	6.927	3.121	9.481
$\lambda \Sigma^{(\infty)}$	7.032	3.170	9.616

considered. Similarly, our results differ substantially from the calculations reported in Ref. [14], in which a combination of the correlation potential method (k th order is denoted $\Sigma^{(k)}$) and the time-dependent Hartree-Fock method with Brueckner orbitals is employed. Moreover, the $E1$ matrix elements quoted in Ref. [13] are improved hugely using the SD_{sc} and $SDpT_{sc}$ methods, where the wave functions are scaled to account for the omitted contributions. Large differences in the results obtained before and after scaling the wave functions demand the inclusion of the omitted contributions more accurately. In Ref. [14] too, the final results are quoted using the $\lambda \Sigma^{(k)}$

approach with the scaling parameter λ . Our CCSD method includes more physical effects through its formulation [23–25] and this is also partly justified by the comparison of energies in Table II. We consider results from the CCSD method to be more reliable since it includes all the nonlinear terms within the considered levels of approximations and accounts for the pair-correlation and core-polarization effects to all orders [22]. These nonlinear terms take care of most of the contributions from the triple and quadrupole excitations; more importantly, both the singly and the doubly excited amplitudes see these effects on equal footing. To show the effectiveness of these nonlinear terms, we also evaluate the $E1$ matrix elements considering the same linear form of the RCC terms in Eq. (36) through our CCSD⁽²⁾ method, which naturally appears in the LCCSD method, and the amplitudes from the CCSD method. As reported in Table III, the differences between the results from the LCCSD and those from the CCSD⁽²⁾ methods are quite large. This supports our above assertion. Compared to the results from the MBPT(2) method, the results obtained using the (LCC)SD and MBPT(3) methods from our calculations and in Ref. [13] are smaller but the CCSD values are closer. This could be because of the fact that there are strong cancellations in the correlation effects among the higher order terms. This trend is similar to the calculations of the energies as listed in Table II. We also note that the amount of contributions estimated through the partial triples effects by us and that given in Ref. [13] are very different. This may be owing to the fact that triples effects are incorporated differently in the two works. The differences between the LCCSD and the LCCSDpT results in our calculations are larger than the differences between the CCSD and the CCSDpT results. This means that the partial triples effects change the results of the LCCSD approximation more than those of the CCSD method. In contrast, we find that the differences between the LCCSD and the LCCSDpT^c results are much smaller than the differences between the CCSD and the CCSDpT^c results, implying that large core correlations are incorporated through the nonlinear terms of the RCC theory. We also observe relatively small changes in the results obtained using the CCSD⁽²⁾, CCSD⁽⁴⁾, and CCSD^(∞) approximations. Thus, the roles of the nonlinear terms of the CCSD method are more effective in the determination of the wave functions than the property evaluation. We also observe that both the Breit and the QED corrections are of a decent size for determining the precise value of the $E1$ matrix element of the $5d^2D_{5/2} \rightarrow 6p^2P_{3/2}$ transition. In fact it is interesting to note here that, unlike in the energy calculations, the total sum of the relativistic corrections to the above $E1$ matrix element obtained from the Breit interaction, VP effect, and SE effect is quite different than the total sum when they are estimated considering all these interactions (Breit + QED approach) together in the CCSD method. In the other transitions, these corrections are found to be slight in magnitude.

In order to satisfactorily address the issues related to the inconsistencies between the previously estimated theoretical results for the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs and the experimental values, it is also essential to estimate the uncertainties associated with the $E1$ matrix elements carefully. Obviously, it can be argued that our calculations have uncertainties from three major sources: (a) using a finite basis

TABLE IV. Reduced matrix elements (in a.u.) due to the $E2$ and $M1$ transitions given by different methods. Relativistic corrections and recommended values along with the uncertainties are listed at the bottom. The most accurate calculations are highlighted in boldface font.

Method	$5d_{3/2} \rightarrow 6s_{1/2}$		$5d_{5/2} \rightarrow 6s_{1/2}$:	$5d_{5/2} \rightarrow 5d_{3/2}$	
	$M1$	$E2$	$E2$	$M1$	$E2$
DHF	~ 0.0	43.844	53.707	1.549	44.287
MBPT(2)	3.2×10^{-5}	34.287	42.217	1.549	29.127
LCCSD	8.8×10^{-5}	31.149	38.642	1.547	23.774
LCCSD ₁₃	8.8×10^{-5}	31.165	38.623	1.547	23.782
LCCSDpT ^c	8.8×10^{-5}	31.149	38.642	1.547	23.774
LCCSD[T]	8.9×10^{-5}	30.698	38.153	1.547	23.033
LCCSDpT	8.7×10^{-5}	31.979	39.541	1.548	24.497
LCCSD _{ex}	9.1×10^{-5}	30.053	37.436	1.548	21.741
CCSD ⁽²⁾	7.6×10^{-5}	35.301	43.437	1.547	28.878
CCSD ⁽⁴⁾	2.1×10^{-4}	35.331	43.468	1.551	28.897
CCSD^(∞)	2.2×10^{-4}	34.400	42.441	1.551	29.037
CCSD ₁₃	2.2×10^{-4}	34.416	42.422	1.551	29.045
CCSDpT ^c	2.1×10^{-4}	34.523	42.598	1.551	29.248
CCSD[T]	2.2×10^{-4}	34.172	42.203	1.551	28.620
CCSDpT	2.0×10^{-4}	34.897	42.960	1.551	29.323
CCSD _{ex}	2.2×10^{-4}	34.530	42.600	1.551	29.622
Relativistic corrections					
Breit	~ 0.0	-0.031	-0.047	~ 0.0	-0.105
VP	~ 0.0	-0.004	-0.004	~ 0.0	0.003
SE	~ 0.0	0.019	0.024	~ 0.0	-0.012
Breit + QED	~ 0.0	-0.015	-0.028	~ 0.0	-0.116
Estimated uncertainties					
Basis	~ 0.0	0.106	0.112	~ 0.0	0.289
Triples	2.0×10^{-5}	0.016	0.019	0.001	0.008
Scaling	~ 0.0	0.130	0.159	~ 0.0	0.585
Recommended values					
Recommended	$2.2(2) \times 10^{-4}$	34.385(168)	42.413(195)	1.550(1)	28.921(653)

size, (b) making approximations in the levels of excitations in the RCC theory, and (c) employing an *ab initio* approach for calculating the wave functions. Among these three, the first two sources of uncertainties are quite understandable. To fathom the uncertainty due to the *ab initio* approach, one can follow from Eq. (32) that both the wave-function and the energy-determining equations are coupled. Therefore, uncertainties associated with both solutions either will cancel each other out or will be added up in the final result. If the experimental energy is used in Eq. (32) (which may be treated as a semiempirical approach), then the uncertainty associated with the energy can be removed (assuming that the experimental energy is more precise). We estimate the uncertainties due to the truncated basis size (“Basis”) by carrying out calculations with the high-lying orbitals using the MBPT(2) method, which are neglected in the RCC calculations to circumvent the computational limitations. Uncertainty due to the neglected triples (“Triples”) are accounted for by taking the difference between the CCSD and the CCSD₁₃ results. To estimate the uncertainties due to the *ab initio* calculations (“Scaling”), we consider the differences in the results between the CCSD and the CCSD_{ex} methods. It is worth mentioning here that most of the partial triples effects seen in our calculations are present inherently within the above estimated differences. Therefore, almost all possible major

uncertainties in our CCSD results are taken into account. By accounting for all these uncertainties in quadrature, we list the recommended values of the $E1$ matrix elements towards the end of Table III. The absolute values are given after adding the relativistic corrections to the CCSD results.

It can be assumed that contributions from the forbidden transition probabilities to the estimations of the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs are negligibly small. However, it is necessary to demonstrate exactly how small they are in the scenario where the aim is to investigate inconsistencies among the theoretical and experimental results. For this purpose, we also estimate these quantities explicitly for the lower order $M1$ and $E2$ forbidden channels. We list these forbidden transition amplitudes from the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states in Table IV using the same methods that were employed to calculate the $E1$ matrix elements. We also list the uncertainties to these quantities adopting the procedure described in the previous paragraph. The trends of these matrix elements obtained with different many-body methods are similar to the $E1$ results except for the $M1$ matrix element between the $5d^2D_{3/2} \rightarrow 6s^2S_{1/2}$ transition—which is, nonetheless, found to be negligibly small. The relativistic corrections are also found to be quite small. The recommended values are listed at the end of the table following the same procedure as given for the $E1$ matrix elements in Table III.

TABLE V. Transition wavelengths (in nm) and probabilities (A_{if}^O) due to different decay channels (O s; in s^{-1}) from the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs from various works. Uncertainties are listed within parentheses. We also compare our results with the other available theoretical and experimental values. Only results from the SDpT_{sc} method are quoted from Ref. [13].

Transition $J_i \rightarrow J_f$	λ_{if} (nm) [46]	O	A_{if}^O (s^{-1})		τ_i (ns)		
			This work	Others	This work	Others	Experiment
$5d^2D_{3/2} \rightarrow 6p^2P_{1/2}$	3011.15	$E1$	986229(18209)	804000 [13]	907(16)	981 [13]	909(15) [16]
$5d^2D_{3/2} \rightarrow 6p^2P_{3/2}$	3613.96	$E1$	116015(2341)	94000 [13]		909 [29]	890(90) [26]
$5d^2D_{3/2} \rightarrow 6s^2S_{1/2}$	689.69	$E2$	21.21(20)			1061 [30]	1250(115) [27]
$5d^2D_{3/2} \rightarrow 6s^2S_{1/2}$		$M1$	~ 0			952 [31]	
						970 [32]	
						856 [33]	
$5d^2D_{5/2} \rightarrow 6p^2P_{3/2}$	3490.84	$E1$	787636(17652)	646000	1270(28)	1369 [13]	1281(9) [16]
$5d^2D_{5/2} \rightarrow 6s^2S_{1/2}$	685.08	$E2$	22.24(21)			1283 [29]	1225(12) [17]
$5d^2D_{5/2} \rightarrow 5d^2d_{3/2}$	102469.52	$E2$	~ 0			1434 [30]	890(90) [26]
$5d^2D_{5/2} \rightarrow 5d^2d_{3/2}$		$M1$	~ 0			1370 [31]	1250(115) [27]
						1342 [32]	1260(80) [28]
						1190 [33]	

Using the recommended transition matrix elements listed in Tables III and IV and the experimental wavelengths quoted in Table V from the database of Ref. [46], we determine the transition probabilities due to all the considered channels from the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs. These values are listed in Table V along with their uncertainties and compared with the values due to the $E1$ channel obtained using the SDpT_{sc} procedure from Ref. [13]. We find large differences between the results from the two works. From the total probabilities of these results, we find that the lifetime of the $5d^2D_{3/2}$ state is 907(16) ns, versus the 981 ns reported in Ref. [13]. This is in quite good agreement with the experimental value of 909(15) ns reported in Ref. [16]. Similarly, we obtain the lifetime of the $5d^2D_{5/2}$ state as 1270(28) ns, versus 1369 ns in Ref. [13]. Our result again agrees well with the experimental value of 1281(9) ns reported in Ref. [16]. In Table V, we also list the estimated lifetimes of these states from some of the previous theoretical and other experimental results. Most of these theoretical estimations were carried out using the nonrelativistic theory [29–33]. Nevertheless, theoretically estimated values in Ref. [29] are very close to our values and the experimental results in Ref. [16]. Other theoretical results are far away from our calculations. Also, other experimental values for the lifetimes of the above $5D$ states have large error bars [26–28] except for the $5d^2D_{5/2}$ state, with 1225(12) ns reported in Ref. [17]. This lies outside the range of the error bar of the value reported in Ref. [16]. Since we have overestimated the uncertainties in our theoretical analysis to provide more reliable results, we anticipate that error bars in our calculations will be smaller than what has actually been reported. From this point of view, our results support the experimental values of the lifetimes of the $5D$ states of Cs reported in Ref. [16]. Our

calculations also demonstrate that the branching ratios for an electron to jump from the $5d^2D_{3/2}$ state to the lower $6p^2P_{1/2}$ state is about 90%, while that to jump to the $6p^2P_{3/2}$ state is about 10%. On the other hand, an electron can jump from the $5d^2D_{5/2}$ to the $6p^2P_{3/2}$ state with almost 100% probability.

V. CONCLUSION

We have employed a variety of relativistic many-body methods, mostly in the CC theory framework, to calculate the energies of the first five low-lying states and the transition matrix elements due to both allowed and forbidden decay channels of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states in Cs. Trends of the results from these methods are discussed and the importance of considering the nonlinear terms in the CC methods for accurate determination of the matrix elements is highlighted. Corrections due to both the Breit interaction and lower order QED effects in these quantities are demonstrated explicitly. Earlier reported inconsistencies between the theoretical and the experimental values of the lifetimes of the above two states seem to be resolved. Branching ratios due to various channels are also given. Though the forbidden transition probabilities are found to be extremely low, our calculated line strengths due to these transitions could be quite useful if the proposed measurements of PNC effects in the $6s^2S_{1/2} \rightarrow 5d^2D_{3/2,5/2}$ transitions in Cs take place in the future.

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