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# Forbidden M1 and E2 transitions in monovalent atoms and ions

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We carried out a systematic high-precision relativistic study of the forbidden magnetic-dipole and electric-quadrupole transitions in  $\text{Ca}^+$ ,  $\text{Rb}$ ,  $\text{Sr}^+$ ,  $\text{Cs}$ ,  $\text{Ba}^+$ ,  $\text{Fr}$ ,  $\text{Ra}^+$ ,  $\text{Ac}^{2+}$  and  $\text{Th}^{3+}$ . This work is motivated by the importance of these transitions for tests of fundamental physics and precision measurements. The relative importance of the relativistic, correlation, Breit correction and contributions of negative-energy states is investigated. Recommended values of reduced matrix elements are presented together with their uncertainties. The matrix elements and resulting lifetimes are compared with other theoretical values and with experiment where available.

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## I. INTRODUCTION

Forbidden transitions have been of much interest in recent years due to their applications in optical clocks [1], tests of fundamental physics [2–7] and quantum information [8]. These applications require long-lived metastable states and, therefore, knowledge of their atomic properties including various multipolar transition rates and branching ratios. While many accurate measurements of the electric-dipole matrix elements exist, there are much fewer precision benchmarks for the M1 and E2 transitions.

The interest in forbidden transitions is further motivated by the emergence of the highly charged ions (HCI) as potential candidates for the development of ultra-precise atomic clocks and tests of variation of fundamental constants [3–5, 9, 10]. Highly charged ions with optical transitions suitable for metrology exhibit a particularly rich variety of low-lying multipolar transitions, even including the metastable levels that can decay only via the M3 decay channel. Until recently, the HCI proposals remained a theoretical possibility, but the first proof-of-principle demonstration of sympathetic cooling of  $\text{Ar}^{13+}$  with laser cooled  $\text{Be}^+$  [11] paved the way toward the experimental realization of the HCI clock proposals. The experimental work toward these new applications of HCIs has already started [12] and reliable predictions of transition properties are urgently needed. While this paper deals with ions of lower degree of ionization, up to  $\text{Th}^{3+}$ , the general conclusions concerning the computational accuracy and the importance of various contributions are also applicable for HCI with a few valence electrons.

We consider examples of the forbidden transitions in  $\text{Rb}$ ,  $\text{Cs}$  and  $\text{Fr}$  alkali-metal atoms and monovalent  $\text{Ca}^+$ ,  $\text{Sr}^+$ ,  $\text{Ba}^+$ ,  $\text{Ra}^+$ ,  $\text{Ac}^{2+}$  and  $\text{Th}^{3+}$  ions with similar electronic structure owing to their particular interest in the applications described above as well as the availability of some experimental measurements. M1 transitions in  $\text{Rb}$ ,  $\text{Cs}$ ,  $\text{Ba}^+$ ,  $\text{Fr}$  and  $\text{Fr}$ -like ions are of particular interest due to studies of parity violation [6, 7, 13–15]. The M1 and E2 transitions in  $\text{Rb}$ ,  $\text{Cs}$ ,  $\text{Ba}^+$ ,  $\text{Yb}^+$ ,  $\text{Ra}^+$ ,  $\text{Ac}^{2+}$  and  $\text{Th}^{3+}$  ions were recently studied by Gossel et al. [16], raising the issue of the accuracy of the M1 transition matrix elements.

The goals of parity violation studies with heavy atoms are to test the Standard Model of particle physics and to study the weak interaction inside the nucleus. In addition, atomic parity violation is uniquely sensitive to possible “dark forces” which are motivated by the intriguing possibility of a “dark sector” extension to the Standard Model [17].

The most accurate, to 0.3%, atomic parity violation measurement was carried out in  $6s - 7s$  transition in  $\text{Cs}$  [2]. The analysis of this experiment in the terms of a comparison with the standard model, which required a theoretical calculation of the parity-violating amplitude, was carried out in [18, 19].

Here, we carry out the calculations of the E2 and M1 matrix elements for monovalent atoms and ions using a form-independent many-body perturbation theory and a relativistic linearized coupled-cluster methods. Previous calculations of the M1 transitions, [20–22], generally assumed that there are no significant corrections beyond the random-phase approximation (RPA). Both of the

methods employed in this work allow us to include corrections beyond RPA. We find that the corrections beyond RPA are large enough to modify the results by a factor of two or more for heavier systems. We also find very strong cancelations of the various corrections beyond RPA for the  $s-s$  (but not the  $s-d$ ) transitions, causing numerical problems in the calculations, associated with the incomplete cancelation of large contributions. We found a way to resolve these problem by using a form-independent perturbation theory as described in the paper. We have also considered the contributions of other effects on the M1 transitions, including the two-body Breit and negative energy state contributions. We have included the study E2 transitions due to their interest of atomic clock and quantum information applications as well as availability of the experimental lifetimes for benchmark tests of the theory. We have conducted a systematic study of our theoretical uncertainties for the E2 matrix elements to provide recommended values for these quantities and compare them with the experimental and other theoretical values.

We start with a review of previous experimental and theoretical studies of the E2 transitions for the systems of interest. The  $s-d$  E2 transition in monovalent ions are used in clock and quantum information applications and lifetimes of  $nd$  states have been the subject of numerous studies described below.

## II. SUMMARY OF PRIOR RESULTS FOR THE E2 TRANSITIONS

**Ca<sup>+</sup>** – Lifetime measurements of the metastable  $3d$  levels of Ca<sup>+</sup> were reported by Knoop *et al.* [23] using the Ca<sup>+</sup> ions stored in a Paul trap. The natural lifetimes were determined to be 1111(46) ms and 994(38) ms, for the  $3d_{3/2}$  and  $3d_{5/2}$  states, respectively, in agreement with previous experiments. An improved measurement of the  $3d_{5/2}$  lifetime, 1168(7) ms, was carried out by Barton *et al.* [24] using quantum jumps of a single cold calcium ion in a linear Paul trap. An experimental and theoretical study of the  $3d$  lifetimes was reported by Kreuter *et al.* [25]. This work introduced a measurement technique based on a high-efficiency quantum state detection after coherent excitation to the  $3d_{5/2}$  state or incoherent shelving in the  $3d_{3/2}$  state and subsequent free, unperturbed spontaneous decay, yielding the value of 1168(9) ms, in agreement with the value reported in Ref. [24]. The lifetime of the  $3d_{3/2}$  state, 1176(11) ms, was measured with a single ion, improving the statistical uncertainty of previous best result by a factor of four. The experimental lifetimes were found to be in excellent agreement with the high-precision *ab initio* all-order calculations, [ $\tau(3d_{3/2})=1196(11)$  ms and  $\tau(3d_{5/2})=1165(11)$  ms], reported in the same work [25]. Sahoo *et al.* [26] used the relativistic coupled-cluster theory to calculate the  $3d$  lifetimes. A large-scale study of the Ca<sup>+</sup> properties, motivated by the development of an atomic clock based on the  $4s-3d_{5/2}$

transition in a Ca<sup>+</sup> single ion, was carried out in [27]. It included the calculation of the blackbody radiation shift of the clock transition, multipole polarizabilities, oscillator strengths, lifetimes, hyperfine constants and excitation energies.

**Sr<sup>+</sup>** – A lifetime measurement of the metastable  $4d_{3/2}$  level in Sr<sup>+</sup> was carried out by Mannervik *et al.* [28] using optical pumping of a stored ion beam. Collinear laser excitation in the storage ring transferred the main part of the ion beam into the metastable  $4d_{3/2}$  level. Subsequent observation of the forbidden electric quadrupole transition to the ground state yielded information about the radiative lifetime of the metastable state, 435(4) ms. The lifetimes of the  $4d$  levels were determined both experimentally and theoretically by Biemont *et al.* [29]. The experiment was performed at an ion storage ring utilizing collinear laser excitation. The calculation was performed by the Hartree-Fock method including relativistic effects and core polarization. The  $4d_{5/2}$  lifetime was measured to be 390(1.6) ms with a single laser-cooled, trapped ion by Letchumanan *et al.* [30] using Dehmelt's electron shelving method to monitor the ions electronic state. Sahoo *et al.* [26] used the relativistic coupled-cluster theory to calculate the  $4d$  lifetimes. A systematic study of Sr<sup>+</sup> atomic properties was carried out in [31] motivated by the development of the Sr<sup>+</sup> clock and the need for the evaluation of the blackbody radiation shift of the clock transitions. Safronova [31] used the relativistic linearized coupled-cluster approach, which included single, double and partial triple excitations, to obtain 441(3) ms and 394(2) ms for the lifetimes of the  $4d_{3/2}$  and  $4d_{5/2}$  states, respectively, in excellent agreement with the experimental values [29, 30].

**Ba<sup>+</sup>** – Lifetimes of the  $5d$  states of Ba<sup>+</sup> are much longer than the corresponding values in Ca<sup>+</sup> and Sr<sup>+</sup>, making their accurate measurement particularly difficult. A single Ba<sup>+</sup> atom was confined in a radio-frequency ion trap and cooled by near-resonant laser light by Madej and Sankey [32]. A measurement of quantum-jump distributions together with careful measurements of the absolute partial pressures of all residual gas species enabled accurate measurements of the quenched  $5d_{5/2}$  lifetime as a function of quenching gas pressure,  $34.5 \pm 3.5$  s [32]. The measurement of the Ba<sup>+</sup>  $5d_{3/2}$  lifetime was carried out by Nagourney and Dehmelt [33] using a single trapped Ba<sup>+</sup> ion in ultrahigh vacuum. The collisional quenching was found insignificant in the experiment, but there were indications of a non-negligible fine-structure mixing effect [33]. The resulting value of  $79.8 \pm 4.6$  s resolved the discrepancy existing at that time. Laser-probing measurements and calculations of lifetimes of the  $5d$  levels were reported by Gurell *et al.* [34]. The lifetimes,  $89.4 \pm 15.6$  s for the  $5d_{3/2}$  level and  $32.0 \pm 4.6$  s for the  $5d_{5/2}$  level, were measured in a beam-laser experiment performed at the ion storage ring CRYRING. The electric-quadrupole transition amplitudes for Ba<sup>+</sup> were evaluated by Gopakumar *et al.* [35] using the relativistic coupled-cluster method, giving the lifetimes of the  $5d_{3/2}$

TABLE I: Recommended values of the reduced electric-quadrupole matrix elements in atomic units. Dirac-Fock DF, third-order many-body perturbation theory MBPT3 and all-order SD are listed. Final recommended values are given in the “Final” column. The relative uncertainties of the final values are given in percent. The rows labeled NBR and BR contain results excluding and including the Breit interaction, respectively. Absolute values are given.

		Transition	DF	MBPT3	SD	Final	(%)	Transition	DF	MBPT3	SD	Final	(%)
Th <sup>3+</sup>	NBr	$7s - 6d_{3/2}$	7.781	6.918	7.063	7.110	0.66	$7s - 6d_{5/2}$	10.008	8.986	9.153	9.211	0.64
	Br	$7s - 6d_{3/2}$	7.781	6.917	7.062	7.109	0.66	$7s - 6d_{5/2}$	10.002	8.979	9.145	9.204	0.64
Ac <sup>2+</sup>	NBr	$7s - 6d_{3/2}$	10.682	9.218	9.515	9.585	0.28	$7s - 6d_{5/2}$	13.655	11.956	12.281	12.366	0.22
	Br	$7s - 6d_{3/2}$	10.679	9.216	9.512	9.585	0.25	$7s - 6d_{5/2}$	13.644	11.944	12.270	12.362	0.15
Ra <sup>+</sup>	NBr	$7s - 6d_{3/2}$	17.263	13.744	14.587	14.736	0.81	$7s - 6d_{5/2}$	21.771	17.802	18.689	18.859	0.70
	Br	$7s - 6d_{3/2}$	17.252	13.734	14.578	14.737	0.74	$7s - 6d_{5/2}$	21.749	17.778	18.667	18.855	0.61
Fr	NBr	$7s - 6d_{3/2}$	43.096	30.292	31.976	33.427	0.58	$7s - 6d_{5/2}$	52.740	37.632	40.017	41.582	0.43
	Br	$7s - 6d_{3/2}$	43.092	30.241	31.937	33.431	0.59	$7s - 6d_{5/2}$	52.729	37.567	39.963	41.582	0.54
Cs	NBr	$6s - 5d_{3/2}$	43.846	30.815	31.548	33.612	0.83	$6s - 5d_{5/2}$	53.712	38.087	39.147	41.464	0.57
	Br	$6s - 5d_{3/2}$	43.830	30.763	31.505	33.620	1.01	$6s - 5d_{5/2}$	53.686	38.013	39.082	41.515	0.85
Ba <sup>+</sup>	NBr	$6s - 5d_{3/2}$	14.763	11.821	12.498	12.627	0.90	$6s - 5d_{5/2}$	18.384	14.863	15.651	15.809	0.85
	Br	$6s - 5d_{3/2}$	14.753	11.813	12.489	12.627	0.83	$6s - 5d_{5/2}$	18.362	14.844	15.632	15.800	0.79
Rb	NBr	$5s - 4d_{3/2}$	38.896	31.793	32.444	32.943	0.42	$5s - 4d_{5/2}$	47.636	38.945	39.755	40.367	0.42
	Br	$5s - 4d_{3/2}$	38.901	31.788	32.156	32.883	0.24	$5s - 4d_{5/2}$	47.642	38.938	39.414	40.295	0.24
Sr <sup>+</sup>	NBr	$5s - 4d_{3/2}$	12.968	10.588	11.010	11.133	0.35	$5s - 4d_{5/2}$	15.972	13.100	13.602	13.747	0.37
	Br	$5s - 4d_{3/2}$	12.961	10.581	11.003	11.133	0.30	$5s - 4d_{5/2}$	15.957	13.088	13.588	13.745	0.29
Ca <sup>+</sup>	NBr	$4s - 3d_{3/2}$	9.767	7.407	7.788	7.945	0.48	$4s - 3d_{5/2}$	11.978	9.099	9.561	9.750	0.47
	Br	$4s - 3d_{3/2}$	9.761	7.401	7.782	7.945	0.47	$4s - 3d_{5/2}$	11.967	9.088	9.552	9.750	0.47

and  $5d_{5/2}$  levels equal to 81.4 s and 36.5 s, respectively. Sahoo *et al.* [26] used the relativistic coupled-cluster theory to obtain 80.0(7) s and 29.9(3) s for these levels, respectively, followed by another calculation of the same group [36]. Reduced electric-quadrupole matrix elements were calculated using both many-body perturbation theory and the all-order method including single, double and partial triple excitations by Safronova [37]. The resulting lifetimes, 81.5(1.3) s and 30.3(0.4) s for the  $5d_{3/2}$  and  $5d_{5/2}$  levels, respectively, were found to be in good agreement with the measured values [34].

**Cs** - While in alkaline-earth metal ions, the first  $nd$  levels are metastable, this is not the case in neutral alkali-metal atoms, where the first  $np$  levels are below the  $nd$  levels and the  $E1$  decay of the  $nd$  levels is allowed. Clab and Nayfen [38] measured the transition probability of the electric quadrupole  $6s - 5d$  transition Cs,  $21 \pm 1.5 \text{ s}^{-1}$ , by two-photon ionization of the ground  $6s$  state, using the  $5d$  as an intermediate state. Previous measurements of this quantity yielded conflicting results. The authors of [38] noted that their measurement was in agreement with a laser absorption-fluorescence measurement and in disagreement with the results of anomalous dispersion, emission, and electron impact techniques.

**Fr and Fr-like ions** - Theoretical studies of the E2  $6d - 7s$  transition rates were carried out in [39]. Safronova *et al.* [40] calculated reduced matrix elements of the E2  $6d - 7s$  transitions in Fr-like Ra and Ac ions using the relativistic linearized coupled-cluster method.

The M1 and E2 transitions in Rb, Cs, Ba<sup>+</sup>, Yb<sup>+</sup>, Ra<sup>+</sup>, Ac<sup>2+</sup> and Th<sup>3+</sup> ions were studied by Gossel *et al.* [16].

TABLE II: Recommended values of the reduced electric-quadrupole matrix elements (in a.u.) are compared with experimental measurements and other theoretical values.

	Transition	Present	Expt.	Theory
Th <sup>3+</sup>	$7s - 6d_{3/2}$	7.110(47)		7.10 [16]
Ac <sup>2+</sup>	$7s - 6d_{3/2}$	9.585(27)		9.58 [16]
Ra <sup>+</sup>	$7s - 6d_{3/2}$	14.74(12)		14.77 [16]
Fr	$7s - 6d_{3/2}$	33.43(19)		35.96(60) [39]
				33.59 [16]
Cs	$6s - 5d_{3/2}$	33.61(28)	34.2(1.2) [38]	33.60 [16]
Ba <sup>+</sup>	$6s - 5d_{3/2}$	12.63(11)	12(1) [34]	12.74 [36]
			12.76(35) [33]	12.63 [35]
Rb	$5s - 4d_{3/2}$	32.94(14)		33.42 [16]
Sr <sup>+</sup>	$5s - 4d_{3/2}$	11.13(39)	11.21(5) [28]	11.33(10) [26]
			11.21(5) [29]	11.13(4) [31]
Ca <sup>+</sup>	$4s - 3d_{3/2}$	7.94(4)	8.01(4) [25]	7.94(4) [25]
			7.92(3) [24]	7.97(2) [26]

### III. ELECTRIC-QUADRUPOLE TRANSITIONS

For electric-quadrupole transitions, we carried out all calculations using four different variants of the linearized coupled-cluster (all-order) method. A review of the all-order method, which involves summing series of dominant many-body perturbation terms to all orders, is given in [41]. In the single-double (SD) all-order approach, single and double excitations of the Dirac-Fock orbitals are included and the SD state vector of a monovalent atom

in state  $v$ :

$$|\Psi_v\rangle = \left[ 1 + \sum_{ma} \rho_{ma} a_m^\dagger a_a + \frac{1}{2} \sum_{mnab} \rho_{mnab} a_m^\dagger a_n^\dagger a_b a_a + \sum_{m \neq v} \rho_{mv} a_m^\dagger a_v + \sum_{mna} \rho_{mnva} a_m^\dagger a_n^\dagger a_a a_v \right] |\Psi_v^{(0)}\rangle, \quad (1)$$

where  $|\Psi_v^{(0)}\rangle$  is the lowest-order atomic state vector and  $a_i^\dagger, a_j$  are creation and annihilation operators. The quantities  $\rho_{ma}, \rho_{mv}$  are single-excitation coefficients for core and valence electrons and  $\rho_{mnab}$  and  $\rho_{mnva}$  are double-excitation coefficients for core and valence electrons, respectively. In Eq. (1), the indices  $m$  and  $n$  range over all possible virtual states while indices  $a$  and  $b$  range over all occupied core states. The single, double, partial-triple (SDpT) method also includes classes of the triple excitations.

In either SD or SDpT all-order method, the matrix elements of any one-body operators, such as M1 and E2,

$$Z = \sum_{ij} z_{ij} a_i^\dagger a_j, \quad (2)$$

are obtained as

$$Z_{wv} = \frac{\langle \Psi_w | Z | \Psi_v \rangle}{\sqrt{\langle \Psi_v | \Psi_v \rangle \langle \Psi_w | \Psi_w \rangle}}, \quad (3)$$

where  $|\Psi_v\rangle$  and  $|\Psi_w\rangle$  are given by the expansion (1). The numerator of Eq. (3) consists of the sum of the lowest-order DF matrix element  $z_{wv}$  and twenty other terms that are linear or quadratic functions of the excitation coefficients  $\rho_{mv}, \rho_{ma}, \rho_{mnva}$ , and  $\rho_{mnab}$ .

The largest terms are frequently

$$Z^{(a)} = \sum_{ma} z_{am} \tilde{\rho}_{wmva} + \sum_{ma} z_{ma} \tilde{\rho}_{vmwa}^*, \quad (4)$$

$$Z^{(c)} = \sum_m z_{wm} \rho_{mv} + \sum_m z_{mv} \rho_{mw}^*. \quad (5)$$

The first of these terms  $Z^{(a)}$  is associated with the random-phase approximation (RPA) corrections, while the second  $Z^{(c)}$  is associated with the Brueckner-orbital corrections; however, there is not a one-to-one correspondence to the many-body classification of corrections to matrix elements.

Omitted higher excitations can also be estimated by the scaling procedure described in [41], which corrects the  $\rho_{mv}$  excitation coefficients and the corresponding terms containing these quantities in Eq. (3), such as term (c). The scaling procedure can be applied to either SD or SDpT approximations. The resulting values are labeled with the subscript  $sc$ ,  $SD_{sc}$  and  $SDpT_{sc}$ . Comparing values obtained in different approximations, *ab initio* SD and SDpT and scaled SD and SDpT allows us to evaluate the uncertainty of the calculations in the cases where the contributions that can be corrected by scaling are

TABLE III: Lifetimes  $\tau$  of the  $nd$  states in  $\text{Ca}^+, \text{Sr}^+, \text{Ba}^+, \text{Ra}^+$  and  $\text{Ac}^{2+}$  in seconds.

Ion	State	Present	Theory	Experiment
$\text{Ca}^+$	$3d_{3/2}$	1.194(11)	0.98 [42]	1.111(46) [23]
			1.271 [43]	1.17(5) [44]
			1.16 [45]	1.20(1) [24]
			1.080 [46]	1.176(11) [25]
			1.196(11) [25]	
			1.185(7) [26]	
$\text{Sr}^+$	$4d_{3/2}$	0.437(14)	0.454 [43]	0.435(4) [28]
			0.422 [29]	0.435(4) [29]
			0.426(7) [26]	0.455(29) [29]
			0.441(3) [47]	
$\text{Ba}^+$	$5d_{3/2}$	81.4(1.4)	83.7 [43]	79.8(4.6) [33]
			81.5 [48]	89(16) [34]
			81.4 [35]	
			80.1(7) [26]	
			82.0 [34]	
			81.5(1.2) [49]	
$\text{Ra}^+$	$6d_{3/2}$	0.6382(94)	0.638(10) [50]	
			0.627(4) [51]	
			0.642 [48]	
			0.642 [15]	
$\text{Ac}^{2+}$	$6d_{3/2}$	$1.171(6) \times 10^6$	$1.19 \times 10^6$ [15]	
$\text{Ca}^+$	$3d_{5/2}$	1.163(11)	0.95 [42]	0.994(38) [23]
			1.236 [43]	1.064(17) [52]
			1.14 [45]	0.969(21) [53]
			1.045 [46]	1.09(5) [44]
			1.165(11) [25]	1.100(18) [54]
			1.110(9) [26]	1.168(7) [24]
$\text{Sr}^+$	$4d_{5/2}$	0.3945(22)	0.405 [43]	0.372(25) [56]
			0.384 [29]	0.408(22) [29]
			0.357(12) [26]	0.3908(16) [30]
			0.394(3) [47]	
			37.2 [43]	32(5) [57]
			30.3 [48]	34.5(3.5) [32]
$\text{Ba}^+$	$5d_{5/2}$	30.34(48)	36.5 [35]	32.0(4.6) [34]
			29.9(3) [26]	31.2(09) [58]
			31.6 [34]	
			30.4(4) [49]	
			0.303(4) [50]	0.232(4) [59]
			0.297(4) [51]	0.232(4) [60]
$\text{Ac}^{2+}$	$6d_{5/2}$	2.326(34)	0.302 [48]	

dominant. We find that this condition is satisfied for the E2 transitions considered in the present work, where term c given by Eq. (5) strongly dominates.

In Table I, we list our recommended values for the  $s-d$  E2 reduced matrix elements in Fr and Fr-like ions, Cs,  $\text{Ba}^+, \text{Rb}, \text{Sr}^+$  and  $\text{Ca}^+$ . The absolute values are

TABLE IV: The M1 matrix elements evaluated in the second-order RMBPT approximation. The lowest-order matrix elements without and with retardation, are listed in columns labeled “DF” and “DF(Ret)”. The second order Coulomb and Breit contributions are listed in the “Cl” and “Br<sub>pos</sub>” columns. The second-order Breit correction calculated with the inclusion of the negative energy (NEG) contributions is given in next column, Br<sub>neg</sub>. The final second-order results MBPT2=Cl+Br<sub>pos</sub>+Br<sub>neg</sub> are listed in the last column. Units:  $10^5 \mu_B$ .

	Transition	DF	DF(ret)	Cl	Br <sub>pos</sub>	Br <sub>neg</sub>	MBPT2
Th <sup>3+</sup>	7s – 8s <sub>1/2</sub>	14.44	11.78	-127.9	-0.232	3.177	-125.1
Ac <sup>2+</sup>	7s – 8s <sub>1/2</sub>	10.07	8.336	-130.1	-0.136	2.304	-128.1
Ra <sup>+</sup>	7s – 8s <sub>1/2</sub>	6.085	5.141	-129.8	0.142	1.252	-128.5
Fr	7s – 8s <sub>1/2</sub>	-2.559	-2.229	56.82	-0.277	-0.353	56.22
Cs	6s – 7s <sub>1/2</sub>	1.952	1.631	-5.001	0.236	0.171	-4.617
Ba <sup>+</sup>	6s – 7s <sub>1/2</sub>	4.952	4.021	-12.710	0.140	0.821	-11.80
Rb	5s – 6s <sub>1/2</sub>	1.824	1.479	0.288	0.207	0.116	0.588
Sr <sup>+</sup>	5s – 6s <sub>1/2</sub>	4.800	3.784	-2.123	0.057	0.730	-1.390
Ca <sup>+</sup>	4s – 5s <sub>1/2</sub>	-4.395	-3.308	-0.063	-0.014	-0.530	-0.557
Th <sup>3+</sup>	7s – 6d <sub>3/2</sub>	1.545	1.560	147.3	0.024	-1.127	146.3
Ac <sup>2+</sup>	7s – 6d <sub>3/2</sub>	1.120	1.121	145.2	0.029	-1.266	144.1
Ra <sup>+</sup>	7s – 6d <sub>3/2</sub>	-0.596	-0.641	-39.46	-0.099	1.380	-38.25
Fr	7s – 6d <sub>3/2</sub>	-0.063	0.074	26.53	0.159	-0.971	25.75
Cs	6s – 5d <sub>3/2</sub>	0.094	-0.026	-2.705	-0.091	0.732	-2.089
Ba <sup>+</sup>	6s – 5d <sub>3/2</sub>	-0.551	-0.562	-14.52	0.036	0.736	-13.81
Rb	5s – 4d <sub>3/2</sub>	-0.289	-0.120	-0.731	0.054	-0.505	-1.165
Sr <sup>+</sup>	5s – 4d <sub>3/2</sub>	0.155	0.206	2.612	-0.019	-0.713	1.945
Ca <sup>+</sup>	4s – 3d <sub>3/2</sub>	-0.041	-0.090	-0.347	0.024	0.625	0.237

given in units of  $ea_0^2$ , where  $a_0$  is the Bohr radius and  $e$  is the elementary charge. Results of first-order Dirac-Fock, third-order many-body perturbation theory and the four all-order calculations described above are listed in the columns labeled DF, MBPT3, SD, SDpT, SD<sub>sc</sub> and SDpT<sub>sc</sub>. We also carried out the calculations using form-independent third-order many-body perturbation theory (MBPT3) method introduced in [61, 62]. The lowest-order values, given in the DF column, illustrate the size of the correlation corrections. The difference of the MBPT3 and the all-order results illustrates the size of the higher-order corrections beyond random-phase approximation, which are included to all orders in MBPT3. Final recommended values are given in the “Final” column. The next column gives the absolute uncertainties. The evaluation of the uncertainty of the matrix elements in this approach was described in detail in [63, 64]. The differences of the all-order values for each transition calculated in different approximation were used to estimate uncertainty in the final results based on the algorithm that accounted for the importance of the specific dominant contributions. The column labeled “Unc. %” of Table I gives relative uncertainties of the final values in percent. The uncertainties are small and range from 0.1% to 1%.

We also investigated the effect of the Breit interaction on the E2 matrix elements. Table I lists the results calculated with and without the Breit interaction. The one-body part of the Breit interaction was included in the construction of the finite basis set which was used in all of the all-order calculations. The two-body Breit correction to matrix elements is small as discussed in detail in [65]. The Breit contribution is very small, less than

0.01% for all cases. The relative uncertainties given in the last column of Table I are less than 1%.

In Table II, our recommended values of the reduced electric quadrupole matrix elements are compared with recent theoretical calculations of Ref. [16]. Most of the other theoretical and all of the experimental papers give the results for the lifetimes of the  $nd$  states of ions levels rather than the  $s - d$  matrix elements. For the  $nd_{3/2}$  lifetimes  $\tau$ , the E2 matrix elements  $Z(nd_{3/2} - n's)$  in a.u. may be accurately extracted using

$$Z = \left[ \frac{(2j+1)\lambda^5}{1.11995 \times 10^{18} \tau} \right]^{-1/2},$$

where  $j = 3/2$ ,  $\lambda$  is the wavelength of the  $ns - n'd_{3/2}$  transition in Å and lifetimes  $\tau$  is in seconds. The contribution of the  $ns - n'd_{3/2}$  transitions is negligibly small.

For the  $nd_{5/2}$  states, there is an additional contribution to the lifetime from the  $nd_{5/2} - nd_{3/2}$  M1 transition. In light ions, Ca<sup>+</sup> and Sr<sup>+</sup>, the contribution of this M1 decay channel to the lifetime is very small, but it becomes significant for Ba<sup>+</sup>, with 18% branching ratio, i.e. relative contribution of the M1 rate to the sum of the M1 and the E2 transition rates. Our matrix elements for the  $6s - 5d_{3/2}$  transitions in Cs is in excellent agreement with experimental measurements given in Ref. [38], with the theoretical prediction having much smaller uncertainty. Our values are in agreement with the experiment for alkaline-earth ions within the uncertainties.

Table III gives the comparisons of the present lifetime results with the experiment [23–26, 29, 33, 34, 44, 52–55] and with other theory [15, 26, 28, 29, 35, 42, 42–51]. No

TABLE V: Recommended values of the reduced magnetic M1 dipole matrix elements in  $10^5 \mu_B$ . The first-order and all-order SD values are listed; Final recommended values are given in the “Final” column. RPA includes lowest-order DF results, the third-order MBPT results (MBPT3) includes both DF and RPA results. The results are compared with DF and RPA values from [16].

		Transition	DF	DF [16]	RPA	RPA [16]	MBPT3	SD	Final
Th <sup>3+</sup>	No Breit	$7s - 6d_{3/2}$	1.545		214.4		111.9	121.9	121.9
	Breit	$7s - 6d_{3/2}$	4.394	4.432	216.0	212.2	123.8	123.1	123.1
Ac <sup>2+</sup>	No Breit	$7s - 6d_{3/2}$	1.119		216.3		123.1	129.6	129.6
	Breit	$7s - 6d_{3/2}$	3.513	3.510	214.5	213.6	113.2	130.3	130.3
Ra <sup>+</sup>	No Breit	$7s - 6d_{3/2}$	0.596		213.7		142.5	138.4	138.4
	Breit	$7s - 6d_{3/2}$	2.368	2.401	212.8	210.3	142.5	138.6	138.6
Fr	No Breit	$7s - 6d_{3/2}$	0.063		128.5		146.3	125.9	125.9
	Breit	$7s - 6d_{3/2}$	0.570	0.737	127.9	126.9	145.8	125.6	125.6
Cs	No Breit	$6s - 5d_{3/2}$	0.094		12.70		13.52	13.23	13.23
	Breit	$6s - 5d_{3/2}$	0.429	0.566	12.95	11.98	14.05	13.84	13.84
Ba <sup>+</sup>	No Breit	$6s - 5d_{3/2}$	0.551		22.72		13.54	15.65	15.65
	Breit	$6s - 5d_{3/2}$	2.009	2.006	23.65	22.06	14.86	16.94	16.94
Rb	No Breit	$5s - 4d_{3/2}$	-0.289		1.214		0.849	1.553	1.553
	Breit	$5s - 4d_{3/2}$	0.017	0.245	1.448	1.019	1.238	2.006	2.006
Sr <sup>+</sup>	No Breit	$5s - 4d_{3/2}$	0.155		4.125		1.853	3.380	3.380
	Breit	$5s - 4d_{3/2}$	1.463		5.210		3.193	4.693	4.693
Ca <sup>+</sup>	No Breit	$4s - 3d_{3/2}$	0.041		-0.657		0.040	0.817	0.817
	Breit	$4s - 3d_{3/2}$	1.203		1.708		1.183	2.012	1.973

experimental lifetimes are available for the  $6d$  levels of the Fr-like ions.

In alkali-metal neutral atoms,  $nd$  states are not metastable and E2 or M1 contributions to the lifetimes are negligible.

#### IV. MAGNETIC DIPOLE MATRIX ELEMENTS

The M1 matrix elements for the  $s - s$  and  $s - d$  transitions are much more difficult to calculate accurately than the E2 ones. For the E2 transitions, the correlation contributes at most 25% to the total, while for the M1 transitions the lowest-order values are very small and the final result comes almost entirely from the correlation corrections. The Breit interaction is more significant as well. Moreover, the negative-energy states,  $\varepsilon_i < mc^2$ , may contribute.

The influence of the negative-energy states (NES) on forbidden magnetic-dipole  $s - s$  transitions in alkali-metal atoms was investigated by Savukov *et al.* in Ref. [66]. The NES contributions were significant in almost all cases and, for rubidium, reduced the transition rate by a factor of 8. Derevianko *et al.* [67] derived the leading term in an  $\alpha Z$  expansion for the negative-energy (virtual electron-positron pair) contributions to the transition amplitudes of heliumlike ions, finding a strong dependence on the choice of the zeroth-order Hamiltonian, which defines the negative-energy spectrum. The ratio of negative-energy contributions to the total transition amplitudes for some nonrelativistically forbidden transi-

TABLE VI: Magnetic dipole (M1) matrix elements in units of  $10^5 \mu_B$ . Relative signs of the present results are adjusted so that final matrix elements are positive. RPA includes lowest-order DF results; the third-order MBPT results (MBPT3) include both DF and RPA. The results are compared with DF and RPA values from [16]

		Tran	DF	DF [16]	RPA	RPA [16]	MBPT3
Th <sup>3+</sup>	NBr	$8s - 7s$	-14.44		158.8		64.68
	Br	$8s - 7s$	-15.78	-13.23	155.2	-2549	61.66
Ac <sup>2+</sup>	NBr	$8s - 7s$	-10.07		172.8		86.97
	Br	$8s - 7s$	-11.13	-8.911	169.4	-2390	84.18
Ra <sup>+</sup>	NBr	$8s - 7s$	-6.085		185.3		112.7
	Br	$8s - 7s$	-6.935	-5.744	182.0	185.1	110.0
Fr	NBr	$8s - 7s$	-2.559		177.1		139.9
	Br	$8s - 7s$	-3.000	-2.49	174.4	176.5	137.4
Cs	NBr	$7s - 6s$	-1.952		14.22		12.45
	Br	$7s - 6s$	-2.189	-1.652	13.66	14.13	11.83
Ba <sup>+</sup>	NBr	$7s - 6s$	-4.952		13.24		8.042
	Br	$7s - 6s$	-5.366	-4.050	12.46	13.53	7.257
Rb	NBr	$6s - 5s$	-1.824		1.004		0.859
	Br	$6s - 5s$	-1.998	-1.473	0.740	1.216	0.553
Sr <sup>+</sup>	NBr	$6s - 5s$	4.800		1.706		2.392
	Br	$6s - 5s$	5.099		2.099		2.828
Ca <sup>+</sup>	NBr	$5s - 4s$	4.395		4.400		4.579
	Br	$5s - 4s$	4.570		4.579		4.791

tions was shown to be of order  $1/Z$ . In the particular case of the magnetic-dipole transition  $3^3S_1 - 2^3S_1$ , authors noted that neglecting of negative-energy contributions,

in an otherwise exact no-pair calculation, would lead one to underestimate the decay rate in helium by a factor of 1.5 in calculations using a Hartree basis and by a factor of 2.9 using a Coulomb basis [67].

The contribution from the negative-energy states for the M1 transitions in Be-like ions was studied by Safronova *et al.*[68], demonstrating that the NES contribution scales as  $\alpha^2 Z$  for both Breit and Coulomb interactions. The relative contribution of the NES was about 0.03% for transitions inside the  $2l2l'$  configuration space and 3% for the  $2l_1 2l_2 - 2l_3 3l_4$  transition. Authors concluded that the NES contributions were important for the weakest transitions in a given transition array.

The E1, E2, M1 and M2 transitions in the nickel iso-electronic sequence were investigated by Hamasha *et al.* [69]. The contributions from negative-energy states were included in the second-order E1, M1, E2 and M2 matrix elements. In second-order matrix elements, such contributions arise explicitly from those terms in the sum over states for which  $\varepsilon_i < mc^2$ . The NES contributions drastically change the second-order Breit-Coulomb matrix elements  $B^{(2)}$ . However, the second-order Breit-Coulomb correction contributes only 2-5% to uncoupled M1 matrix elements and, as a result, negative-energy states changed the total values of M1 matrix elements by only a few percent [69].

The contributions from negative-energy states were included in the second-order E1, M1, E2 M2, E3 and M3 matrix elements in [70]. The NES contributions to the second-order Breit-Coulomb matrix elements for the transition from  $3d_{5/2}5d_{3/2}(1)$  state in Ni-like ions weakly increases with  $Z$ , however, the relative NES contribution for this transition decreases with  $Z$  (2% and 0.6% for  $Z = 40$  and  $Z = 90$ , respectively). Ref. [70] noted that the NES contribution for this transition are of the same order as the positive-energy state contribution to the second-order Breit-Coulomb matrix elements causing severe cancelation and drastically reducing the  $B^{(2)}$  values in this case. Therefore, we include the contributions of the NES as well as retardation corrections and correlation effects for the M1 transitions in detail in the second-order MBPT calculation.

In Table IV, we list the M1 magnetic matrix elements evaluated in second-order RMBPT approximation. We employ customary units for reduced matrix elements as given in the National Institute for Science and Technology compendium on Atomic Spectroscopy [71], These units are  $e^2 a_0^2$  for E2 transitions and  $\mu_B$  for M1 transitions.

The lowest-order DF values are evaluated with the relativistic version of the M1 operator without retardation. The DF(Ret) values include retardation. The table illustrates that the retardation corrections are particularly large for the  $s - d$  transition in Rb, Cs and Fr. The second-order Coulomb and Breit contributions are listed in the “Cl” and “Br<sub>pos</sub>” columns. The second-order Breit correction, which includes the negative energy (NEG) contributions, is given in next column, Br<sub>neg</sub>. The final

second-order results MBPT2=Cl+Br<sub>pos</sub>+Br<sub>neg</sub> are listed in the last column. We find that the NES effect on the Coulomb correlation correction is negligible and can be omitted without the loss of accuracy, and it is not shown in the table. The contribution of the NES to the second-order Breit correction is significant, as illustrated by the significant differences of the  $B_{\text{pos}}$  and  $B_{\text{neg}}$  values. However, the table clearly indicates that the Coulomb correlation correction dominates the final values and an accurate calculation of this correction presents a significant challenge. As noted above, the M1 transitions between levels of different electronic configurations are extremely sensitive to the correlation correction, since the lowest-order M1 values are very small and the final result comes almost entirely from the correlation correction.

While it was previously assumed that only RPA corrections contribute significantly to the M1 matrix elements, we find that it is not the case for the transitions studied in this work.

In Table V, we list our values for the M1  $s - d$  reduced matrix elements in units of  $10^5 \mu_B$ . The final results are obtained using the same all-order approach as for the E2 matrix elements. The four variants of the all-order calculations are carried out for the M1 transitions as for the E2 transitions. We also carried out the calculations of the M1 matrix elements using form-independent third-order many-body perturbation theory (MBPT3) method introduced in [61, 62]. The all-order values are taken as final. We find that while using the form of the M1 operator that includes retardation changes the DF values, its effect on the final result is negligible at the present level of accuracy and is omitted in Table V. Results with and without the inclusion of the Breit interaction are listed, with the Breit contribution being more important for the M1 transitions in comparison with the E2 transitions.

Comparing the third-order MBPT3 and RPA results demonstrates that corrections beyond RPA are large for all cases, in particular Fr and Fr-like ions. The MBPT3 classification and formulas for such corrections, which include Brueckner-orbital (BO), structure radiation (SR), and normalization is given in [72]. The form-independent variant of the third-order used here includes further corrections due to replacement of the DF matrix elements by the “dressed” RPA values in all formulas. This approach is discussed in detail in [61, 62]. The all-order SD calculations include all of the third-order and additional higher-order correlation corrections. The comparison of the MBPT3 and all-order SD values demonstrate that the fourth and higher-order contributions are significant for these M1 transitions.

The same calculations are carried out for the  $s - s$  transitions. The results are presented in Table VI, where DF, RPA and the MBPT3 final values are listed. Our M1 values are compared with the theoretical results from Ref. [16] obtained in the DF and RPA approximations. Negative-energy and retardation corrections are omitted, these contributions are smaller than the uncertainty in the correlation corrections as demonstrated in Table IV.

DF energies are used to define  $\omega$  in all RPA calculations. The MBPT3 values are taken as final.

We identified two issues in the calculations of these matrix elements. First, we find that there are significant numerical instabilities in Dirac-Fock computations of the M1  $s-s$  matrix elements (the effect is small for the  $s-d$  case). The DF codes used for generation of the several low-level orbitals do not usually orthonormalize the resulting wave functions, since it is done by subsequent basis set codes. In the relativistic case one expects the accuracy of the M1 radial matrix element to be limited by the size of the overlap matrix integral  $(g_v g_w + f_v f_w) dr$ , where  $g$  and  $f$  are the large and small components of the wave function and  $v$  and  $w$  indicate initial and final electron states. If the  $ns$  orbitals are not orthonormal to a good numerical precision, the respective integral is not numerically stable, leading to spurious errors, generally of a few per cent. This problem does not arise in the present RPA, MPBT3, and all-order calculations since we do all computations with the orthonormalized basis set wave functions. However, it explains the difference with the DF and RPA results of [16], which used DF functions in the RPA calculations. This issue is a potential source of the drastic difference or our RPA values with [16] for the Fr-like Ac and Th ions. The M1 matrix elements for Fr-like Ac and Th ions are not expected to be significantly different from the Fr and Fr-like Ra values and present RPA and final MBPT3 values for Fr-like isoelectronic sequence shows smooth changes.

The second problem with the calculation of the  $s-s$  transitions is a strong cancelation of the large Brueckner-orbital (BO) and structure radiation (SR) corrections. In the all-order case, the BO-type term  $c$  given by Eq.(5) is very large but is strongly canceled by SR-type term

$$Z^{(p)} = \sum_{mnra} z_{mn} \tilde{\rho}_{rmwa}^* \tilde{\rho}_{rnva}. \quad (6)$$

Either of these terms is at least an order of magnitude larger than the RPA. This issue makes the all-order computation of the M1  $s-s$  matrix elements unreliable in its current implementation. Most likely, omission of the triple- and higher-excitations leads to incomplete cancelations and full inclusion of the other high-order corrections, such as those from non-perturbative triple excitations, non-linear terms, and others is needed. An inclusion of the perturbative triples or scaling exacerbates the problem instead of correcting it, since they directly

affect only BO-type terms but not the structural radiation. The  $s-d$  transitions do not present such problems: we find some significant contributions from the non-RPA terms described above but no strong cancelations.

To improve upon the RPA results for the  $s-s$  transition, we use a form-independent third-order MBPT method introduced in [61, 62]. This approach yields electric-dipole transition amplitudes that are equal in the length and velocity forms for transitions in atoms with one valence electron within the framework of relativistic many-body perturbation theory starting from the Dirac-Hartree-Fock approximation. For the M1 transitions, where the matrix elements are in velocity form, such an approach appears to provide more accurate cancelations of the large BO and SR many-body corrections. Even with the strong cancelations, the remaining corrections are still significant for the  $s-s$  M1 matrix elements. Further improvement of the theoretical accuracy may be achieved with the development of the form-independent all-order approach.

## V. CONCLUSION

In summary, we carried out a systematic relativistic study of  $s-s$  and  $s-d$  M1 transitions in Fr and Fr-like ions, Cs, Ba<sup>+</sup>, Rb, Fr, Cs, Ba<sup>+</sup>, Rb, SR<sup>+</sup>, and Ca<sup>+</sup> atomic systems. Benchmark comparisons of the  $nd$  lifetimes are carried out. Relativistic, correlation, Breit and negative-energy contributions are studied. The estimated accuracy of the theoretical  $s-d$  E2 matrix elements is very high, better than 1%. and is good for the  $s-d$  M1 matrix elements. A rough estimate of the accuracy of  $s-d$  M1 matrix elements is given by the difference between the SD and MBPT3 values listed in Table IV which can exceed 10%. We find that inclusion of the correction beyond RPA is essential for accurate calculations of the M1 matrix elements considered in this work. New high-precision experimental results are urgently needed for the M1 transitions to test theoretical predictions.

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