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Theoretical study of lifetimes and polarizabilities in Ba$^+$

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The $6s$-np_j (n=6–9) electric-dipole matrix elements and $6s$-nd_j (n=5–7) electric-quadrupole matrix elements in Ba$^+$ are calculated using the relativistic all-order method. The resulting values are used to evaluate ground-state dipole and quadrupole polarizabilities. In addition, the electric-dipole $6p_j$-$5d_{j'}$ matrix elements and magnetic-dipole $5d_{5/2}$-$5d_{3/2}$ matrix element are calculated using the same method in order to determine the lifetimes of the $6p_{1/2}$, $6p_{3/2}$, $5d_{3/2}$, and $5d_{5/2}$ levels. The accuracy of the $6s$-$5d_j$ matrix elements is investigated in detail in order to estimate the uncertainties in the quadrupole polarizability and $5d_j$ lifetime values. The lifetimes of the $5d$ states in Ba$^+$ are extremely long, making precise experiments very difficult. Our final results for dipole and quadrupole ground-state polarizabilities are $\alpha_{E1}=124.15a_0^3$ and $\alpha_{E2}=4182(34)a_0^5$, respectively. The resulting lifetime values are $\tau_{6p_{1/2}}=7.83$ ns, $\tau_{6p_{3/2}}=6.27$ ns, $\tau_{5d_{3/2}}=81.5(1.2)$ s, and $\tau_{5d_{5/2}}=30.3(4)$ s. An extensive comparison with other theoretical and experimental values is carried out for both lifetimes and polarizabilities.

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I. INTRODUCTION

The atomic properties of Ba$^+$ ion are of particular interest owing to the prospects of studying the parity nonconservation (PNC) with a single trapped ion [1]. Progress on the related spectroscopy with a single Ba$^+$ ion is reported in [2,3], and precision measurements of light shifts in a single trapped Ba$^+$ ion have been reported in [4]. The PNC interactions give rise to nonzero amplitudes for transitions that are otherwise forbidden by the parity selection rules, such as the $6s$-$7s$ electric-dipole transition in Cs. The study of parity nonconservation in cesium [5,6] involving both high-precision measurements and several high-precision calculations provided an atomic-physics test of the standard model of electroweak interactions and yielded the first measurement of the nuclear anapole moment (see [7] for a review of studies of fundamental symmetries with heavy atoms). The analysis of the Cs experiment, which required a calculation of the nuclear spin-dependent PNC amplitude, led to constraints on weak nucleon-nucleon coupling constants that are inconsistent with constraints from deep inelastic scattering and other nuclear experiments [8]. More PNC experiments in other atomic systems, such as Ba$^+$, are needed to resolve this issue. The prospects for measuring parity violation in Ba$^+$ have been recently discussed in [3].

Ba$^+$ is also of particular interest for developing optical frequency standard [9] and quantum information processing [10,11] owing to the extremely long lifetimes of $5d$ states. The accuracy of optical frequency standards is limited by the frequency shift in the clock transitions caused by the interaction of the ion with external fields. Therefore, knowledge of atomic properties is needed for the analysis of the ultimate performance of such frequency standard.

Another motivation for study of Ba$^+$ is an excellent opportunity for tests of theoretical and experimental methods, in particular in light of recent measurements of Ba$^+$ atomic properties [2,3,12–15]. Ba$^+$ is a monovalent system allowing for precise theoretical predictions and, in some cases, for evaluation of the theoretical uncertainties that do not directly rely on the comparison with experiment. It is also an excellent testing case for further studies of Ra$^+$ ion, where the correlation corrections are expected to be larger owing to a larger core. A project to measure PNC in a single trapped radium ion was recently started at the KVI, Groningen [16].

In this work, we calculate $6s$-np_j (n=6–9), $6p_j$-$5d_{j'}$ electric-dipole matrix elements, $6s$-nd_j (n=5–9) electric-quadrupole matrix elements, and $5d_{5/2}$-$5d_{3/2}$ magnetic-dipole matrix element in Ba$^+$. This set of matrix elements is needed for accurate calculation of ground-state dipole and quadrupole polarizabilities and lifetimes of the $6p_{1/2}$, $6p_{3/2}$, $5d_{3/2}$, and $5d_{5/2}$ levels. We carefully investigate the uncertainty in our values of $6s$-$5d_j$ matrix elements in order to estimate the uncertainties in the quadrupole polarizability and the $5d_j$ lifetime values. It is particularly important to independently determine these uncertainties because of significant inconsistencies between different measurements of the $5d_{3/2}$ and $5d_{5/2}$ lifetimes [15,17–22]. There are also large discrepancies between experimental determinations of the $5d$-$6s$ quadrupole matrix element from the lifetime experiments and studies of the Rydberg states of barium [12–14]. The experimental values of the ground-state quadrupole polarizability from Refs. [12,14,23] differ by a factor of 2; our value of the quadrupole polarizability is in agreement with Ref. [14]. We note that there are no inconsistencies between the experimental lifetimes [24–26] of the $6p_j$ levels and experimental determinations of the electric-dipole ground-state polarizability [12,14,23]. The experimental values of the electric-dipole polarizability of the Ba$^+$ ion in its ground state [12,14,23] are also in agreement with each other and our theoretical value. Our lifetimes of the $6p_{1/2}$ and $6p_{3/2}$ levels are in agreement with experimental values [24–26] within expected accuracy (1%).

The paper is organized as follows. In Sec. II, we give a short description of the method used for the calculation of the matrix elements. In Sec. III, we discuss the calculation of the electric-dipole polarizability and conduct comparative analysis of the correlation corrections to the ns-np matrix elements in Ba$^+$, Cs, and Ca$^+$. The $6s$-$5d$ quadrupole matrix elements and the ground-state quadrupole polarizability are discussed in Sec. IV, and the lifetimes are discussed in Sec. V. A consistency study of the $5d_j$ lifetime and ground-state quadrupole polarizability measurements in Ba$^+$ is presented in Sec. IV.

II. METHOD

We calculate the reduced multipole matrix elements using the relativistic all-order method [27–29] which is a linearized coupled-cluster method where all single and double excitations of the Dirac-Fock wave function are included to all orders of perturbation theory. The present implementation of the method is suitable for the calculation of matrix elements of any one-body operator; i.e., the calculations of the $E1$, $E2$, and $M1$ matrix elements are carried out in the same way. We refer the reader to the review in [29] and references therein for the detailed description of the all-order method.

Briefly, our starting point is the relativistic no-pair Hamiltonian [30] expressed in second quantization as

$$H=\sum_i \epsilon_i :a_i^\dagger a_i: + \frac{1}{2}\sum_{ijkl} g_{ijkl} :a_i^\dagger a_j^\dagger a_l a_k:, \qquad (1)$$

where $a_i^\dagger$ and a_j are single-particle creation and annihilation operators, respectively, ϵ_i is the Dirac-Fock (DF) energy for the state i, g_{ijkl} are the two-body Coulomb integrals, and :: indicates normal order of the operators with respect to the closed core. The single-double (SD) all-order wave function is written as

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

where $|\Phi_v\rangle$ is the lowest-order wave function taken to be the frozen-core DF wave function of a state v. Indices at the beginning of the alphabet, $a,b,\ldots$, refer to occupied core states; those in the middle of the alphabet, $m,n,\ldots$, refer to excited states; and the index v designates the valence orbital. The all-order equations for the excitation coefficients ρ_{ma}, ρ_{mv}, ρ_{mnab}, and ρ_{mnva} are solved iteratively with a finite basis set, and the correlation energy is used as a convergence parameter. The basis set is defined in a spherical cavity on a nonlinear grid and consists of single-particle basis states which are linear combinations of B-splines [31]. We use a basis set of 50 splines of order 9 in a spherical cavity of radius 80 a.u. Such cavity size is chosen to accurately represent all orbitals of interest to the present study. The resulting excitation coefficients ρ_{ma}, ρ_{mv}, ρ_{mnab}, and ρ_{mnva} are used to calculate the one-body $E1$, $M1$, and $E2$ matrix elements.

The SD all-order method yielded results for the primary ns-np_j $E1$ matrix elements of alkali-metal atoms that are in agreement with experiment to 0.1%–0.5% [28]. We note that while the all-order expression for the matrix elements contains 20 terms that are linear or quadratic functions of the excitation coefficients, only two terms are dominant for all matrix elements considered in this work:

$$Z^{(a)} = \sum_{ma} (z_{am}\tilde{\rho}_{wmva} + z_{ma}\tilde{\rho}^{*}_{vmwa}) \qquad (3)$$

and

$$Z^{(c)} = \sum_{m} (z_{wm}\rho_{mv} + z_{mv}\rho^{*}_{mw}), \qquad (4)$$

where $\tilde{\rho}_{mnab}=\rho_{mnab}-\rho_{nmab}$ and z_{wv} are lowest-order matrix elements of the corresponding operator. In the case of the electric-quadrupole transitions studied in this work, the second term $Z^{(c)}$ is overwhelmingly (by an order of magnitude) larger than any other term. In such cases, it was found necessary to include at least partially triple excitations into the wave function

$$|\Psi_v^{\mathrm{SDpT}}\rangle = |\Psi_v^{\mathrm{SD}}\rangle + \frac{1}{6}\sum_{mnrab} \rho_{mnrvab} a_m^\dagger a_n^\dagger a_r^\dagger a_b a_a a_v |\Phi_v\rangle \qquad (5)$$

and to correct the single excitation coefficient ρ_{mv} equation for the effect of triple excitations [28,32–34]. We have conducted such a calculation for the $6s$-$5d_j$, $6s$-$6d_j$, and $6s$-$7d_j$ electric-quadrupole matrix elements and refer to the corresponding results as SDpT values (i.e., including all single, double, and partial triple excitations).

We note that such approach works poorly when terms $Z^{(a)}$ and $Z^{(c)}$ are of similar order of magnitude (such as all $E1$ transition considered here) owing most likely to cancellation of high-order corrections to terms $Z^{(a)}$ and $Z^{(c)}$. The term $Z^{(a)}$ is not directly corrected for triple excitations in the SDpT extension of the method leading to consistent treatment of the higher-order correlations only when the second term is overwhelmingly dominant. We refer the reader to Ref. [35] for a detailed discussion of triple excitations. The results of the matrix element calculation are discussed in the following sections.

III. Ba$^+$ GROUND-STATE DIPOLE POLARIZABILITY

The ground-state dipole or quadrupole polarizability can be represented as a sum of the valence polarizability α_v and the polarizability of the ionic core α_{core} [28]. The calculation of the core polarizability assumes allowed excitations to any excited state including the valence shell, which requires the introduction of the small counter terms α_{vc} to subtract out 1/2 of the contribution corresponding to the $6s$ shell excitation [28]. The core polarizabilities have been calculated in random-phase approximation (RPA) in Ref. [36]. The accuracy of the RPA values is expected to be on the order of 5% [34]. We calculated the α_{vc} term the in the RPA for consistency with the α_{core} value. The valence dipole polarizability for the $6s$ state of Ba$^+$ is calculated as a sum over states:

$$\alpha_{v,E1} = \frac{1}{3}\sum_n \left(\frac{|\langle 6s\|d\|np_{1/2}\rangle|^2}{E_{np_{1/2}}-E_{6s}} + \frac{|\langle 6s\|d\|np_{3/2}\rangle|^2}{E_{np_{3/2}}-E_{6s}} \right). \qquad (6)$$

The sum over the principal quantum number n in Eq. (6) converges very rapidly, and very few first terms have to be calculated to high precision. In this work, we use SD all-order matrix elements and experimental energies for terms with $n=6-9$ and evaluate the remainder α_{tail} in the Dirac-

TABLE I. Contributions to the ground-state $6s$ scalar dipole polarizability α_{E1} in Ba$^+$ in units of a_0^3. Comparison with experiment and other calculations. The absolute values of corresponding SD all-order reduced electric-dipole matrix elements d (in a.u.) are also given.

Contribution	d	α_{E1}
$6s$-$6p_{1/2}$	3.3357	40.18
$6s$-$6p_{3/2}$	4.7065	73.82
$6s$-$7p_{1/2}$	0.0621	0.06
$6s$-$7p_{3/2}$	0.0868	0.01
α_{tail}		0.03
α_{core}		10.61
α_{vc}		−0.51
Total		124.15
Expt. [14]		123.88(5)
Expt. [23]		125.5(10)
Theory [41]		123.07
Theory [42]		126.2
Theory [43]		124.7

Fock approximation. The contributions to the dipole polarizability are summarized in Table I. We also list the absolute values of corresponding SD all-order reduced electric-dipole matrix elements d. The contribution of the terms with $n=6$ is overwhelmingly dominant. Therefore, the uncertainty in our calculation of the dipole polarizability is dominated by the uncertainties in the $6s$-$6p_{1/2}$ and $6s$-$6p_{3/2}$ matrix elements.

To study the uncertainty in these values, we investigate the importance of the contributions from various correlation correction terms and the overall size of the correlation correction. The contributions to the $6s$-$6p_{1/2}$ matrix element are summarized in Table II. The breakdown of the contributions to the $6s$-$6p_{3/2}$ matrix element is essentially the same, and we do not list it here. We also give the breakdown of the correlation correction for the same transition in Cs and $4s$-$4p_{1/2}$ transition in Ca$^+$. Cs values are taken from Ref. [37]. The final Ca$^+$ value has been published in Ref. [38]. As we noted in Sec. II, only two terms give large contributions to the correlation correction. While there are some cancellations in the other terms, all them are at least an order of magnitude smaller. Unfortunately, there is no straightforward way to evaluate the uncertainty in the $Z^{(a)}$ term (as we show in the later section, it can be done for $Z^{(c)}$). Therefore, we cannot make an uncertainty estimate that is independent on experimental observations. However, we note that Cs $6s$-$6p_j$ transitions are extremely well studied by a number of different experimental approaches (see, for example, [39] and references therein), and all-order SD data are in agreement with Cs experimental values to 0.2%–0.4% [28]. The breakdown of terms for Ba$^+$ is slightly different than for Cs, but is very similar to Ca$^+$. As expected, the size of the correlations is larger in Ba$^+$ than in Ca$^+$. Unfortunately, there is only one high-precision measurement of the $4p_j$ Ca$^+$ lifetimes [40] that is in significant (3%) disagreement with high-precision theoretical results. Similar discrepancies existed for the alkali-metal atom measurements done with the same technique and later experiments confirmed the theory values. We refer the reader to Ref. [38] for a more detailed discussion of this issue. It would have been very interesting to see the $4p$ lifetimes in Ca$^+$ remeasured to resolve this issue. Based on the similar size of the correlation corrections for Cs and Ba$^+$, we expect similar accuracy of our data (on the order of 0.5%). Therefore, the resulting accuracy of our dipole polarizability is expected to be on the order of 1%. We find that our value is in excellent agreement with both experimental values [14,23] when our estimated uncertainty is taken into account. Our results are in good agreement with other theoretical calculations [41–43]. We also note that the $\langle 6s\|d\|6p\rangle$ matrix element has been recently extracted from the K splittings of the bound $6snl$ states in Ref. [13], and the resulting value $\langle 6s\|d\|6p\rangle=4.03(12)$ is in excellent agreement with our result $\langle 6s\|d\|6p\rangle=4.08$ (normalized spherical harmonics C_1 is factored out here for comparison).

TABLE II. Contributions of different terms to the Ba$^+$, Ca$^+$, and Cs ns-$np_{1/2}$ reduced matrix elements in a.u.

Contribution	Ba$^+$	Cs [37]	Ca$^+$
	$6s$-$6p_{1/2}$	$6s$-$6p_{1/2}$	$4s$-$4p_{1/2}$
DF	3.891	5.278	3.201
$Z^{(a)}$	−0.387	−0.334	−0.200
$Z^{(c)}$	−0.209	−0.485	−0.120
Other	0.041	0.019	0.016
Total	3.336	4.478	2.898
Correlation	16.6%	17.9%	10.5%

IV. Ba$^+$ GROUND-STATE QUADRUPOLE POLARIZABILITY

The valence part of the quadrupole polarizability is given in the sum-over-states approach by

$$\alpha_{v,E2}=\frac{1}{5}\sum_n\left(\frac{|\langle 6s\|Q\|nd_{3/2}\rangle|^2}{E_{nd_{3/2}}-E_{6s}}+\frac{|\langle 6s\|Q\|nd_{5/2}\rangle|^2}{E_{nd_{5/2}}-E_{6s}}\right). \quad (7)$$

The RPA core value [36] is $46a_0^5$, and the α_{vc} term is negligible. The terms containing the $6s$-$5d_{3/2}$ and $6s$-$5d_{5/2}$ matrix elements give an overwhelmingly dominant contribution to the total values. Therefore, we study these transitions in more detail and evaluate their uncertainties. Unlike the case of the $E1$ transitions considered earlier, the $Z^{(c)}$ term contributes over 90% of the total correlation correction. Therefore, we carried out the calculation using both SD and SDpT approaches described in Sec. II. We also carried out semiempirical scaling in both approximations by multiplying single excitation coefficients ρ_{mv} by the ratio of the "experimental" and corresponding (SD or SDpT) correlation energies [32]. The "experimental" correlation energies are determined as the difference of the total experimental energy and the DF lowest-order values. The calculation of the matrix elements is then repeated with the modified excitation coefficients. The accuracy of such a scaling procedure for similar cases

TABLE III. Absolute values of electric-quadrupole $6s$-$5d_{3/2}$ and $6s$-$5d_{5/2}$ reduced matrix elements in Ba$^+$ calculated in different approximations in a.u. Columns labeled "DF" and "III" are lowest-order Dirac-Fock and third-order many-body perturbation theory (MBPT) values, respectively. The third-order results calculated with maximum number of partial values $l_{max}=6$ and $l_{max}=10$ are given to illustrate the contribution of the higher partial waves. Breit correction is given separately. The all-order *ab initio* results are given in columns labeled "SD" and "SDpT," respectively; these results include contributions from higher partial waves and Breit correction. The corresponding scaled values are listed in columns labeled "SD$_{sc}$" and "SDpT$_{sc}$." The calculation of the uncertainties of the final values is described in detail in text.

Transition	DF	III ($l_{max}=6$)	III ($l_{max}=10$)	Breit	SD	SDpT	SD$_{sc}$	SDpT$_{sc}$	Final
$6s$-$5d_{3/2}$	14.76	11.82	11.75	−0.07	12.42	12.66	12.63	12.59	12.63(9)
$6s$-$5d_{5/2}$	18.38	14.86	14.78	−0.09	15.55	15.84	15.80	15.76	15.80(11)

was discussed in detail in Refs. [33,34,44]. The reasoning for such a scaling procedure in third-order perturbation theory (scaling of the self-energy operator) has been discussed in Ref. [45]. We list SD, SDpT, and the corresponding scaled results (labeled "SD$_{sc}$" and "SDpT$_{sc}$") in Table III. The lowest-order DF results are listed to illustrate the size of the correlation corrections. We demonstrate the size of the two other corrections—the contribution of the higher partial waves and Breit correction—in the same table. The first correction results from the truncation of the partial waves in all sums in all-order calculation at $l_{max}=6$. The all-order calculation with higher number of partial waves is impractical. Therefore, we carry out the third-order MBPT calculation (following Ref. [45]) including all partial waves up to $l_{max}=6$ and $l_{max}=10$ and take the difference of these two values to be the contribution of the omitted partial waves that we add to *ab initio* all-order results. We verified that the contribution of the $l=9-10$ partial waves is very small, justifying the omission of contributions from $l>10$. The Breit correction is calculated as the difference of the third-order results with two different basis sets. The second basis set is generated with taking into account one-body part of the Breit interaction. We note that scaled values should not be corrected for either partial wave truncation error or Breit interaction to avoid possible double-counting of the same effects. We take SD$_{sc}$ values as our final results. The uncertainty of the final values is calculated as follows: the uncertainty in the $Z^{(c)}$ term is evaluated as the spread of the most high-precision values (SD$_{sc}$, *ab initio* SDpT, and SDpT$_{sc}$); the remaining theoretical uncertainty in the Coulomb correlation correction is taken to be the same as the uncertainty in the dominant $Z^{(c)}$ term. We assume 100% uncertainties in the contributions of the higher partial waves and Breit correction. The final uncertainty of the $6s$-$5d_j$ matrix elements (0.7%) is obtained by adding these four uncertainties in quadrature. We note that this procedure for the uncertainty evaluation does not rely on the experimental values with the exception of the experimental energies used for scaling.

The contributions to the ground-state quadrupole polarizability are given in Table IV. While the $n=5$ term is dominant, the contributions of the few next terms are substantial. Therefore, we carry out SD, SDpT, and both scaled calculations for the $6s$-$6d_j$ and $6s$-$7d_j$ matrix elements as well and repeat the uncertainty analysis described above (we omit Breit and higher-partial-wave corrections here since such precise evaluation of the uncertainties is not needed for these transitions). The $6s$-$8d_j$ and $6s$-$9d_j$ matrix elements are calculated in third-order MBPT, and their accuracy is taken to be 2% based on the comparison of the third-order and all-order values of the $6s-7d_j$ matrix elements. The remainder is evaluated in the DF approximation and reduced by 23% based on the comparison of the DF and third-order data for $6s$-$8d_j$ and $6s$-$9d_j$ matrix elements. Its accuracy is correspondingly taken to be 23%.

Our recommended value for the ground-state quadrupole polarizability is in agreement within the corresponding uncertainties with the most recent experimental work [14]. However, our value for the contribution of the $6s$-$5d_j$ transitions to the quadrupole polarizability [3368(34)] differs by about a factor of 2 from the experimental values [12–14] obtained based on the nonadiabatic effects on the Rydberg fine-structure intervals. This issue and the discrepancies in the experimental values of the quadrupole polarizabilities are

TABLE IV. Contributions to the ground-state $6s$ quadrupole polarizability α_{E2} in Ba$^+$ and their uncertainties in units of a_0^5. The absolute values of corresponding all-order reduced electric-quadrupole matrix elements Q (in a.u.) and their uncertainties are also given.

Contribution	Q	α_{E2}
$6s$-$5d_{3/2}$	12.63(9)	1436(20)
$6s$-$6d_{3/2}$	16.83(5)	270(2)
$6s$-$7d_{3/2}$	5.68(5)	23.7(4)
$6s$-$8d_{3/2}$	3.09(6)	6.3(3)
$6s$-$9d_{3/2}$	2.07(4)	2.7(1)
$6s$-$5d_{5/2}$	15.8(1)	1932(27)
$6s$-$6d_{5/2}$	20.30(6)	392(2)
$6s$-$7d_{5/2}$	6.98(6)	35.7(6)
$6s$-$8d_{5/2}$	3.83(8)	9.6(4)
$6s$-$9d_{5/2}$	2.57(5)	4.19(2)
α_{tail}		24(6)
α_{core}		46(2)
Total		4182(34)
Expt. [14]		4420(250)
Expt. [12]		2462(361)
Expt. [23]		2050(100)

addressed in detail in Ref. [14]. We note that these experimental values of the $6s$-$5d_j$ contributions to the quadrupole polarizabilities (1524(8) [14] and 1562(93) [13] in the two most recent studies) are significantly inconsistent with all high-precision calculations of the $5d_j$ lifetimes [15,46–50] carried out by different methods as well as with all experimental lifetime measurements (also carried out by different techniques) [15,17,19–22]. For comparison, the value 1562(93) obtained from the $\langle 6s \| r^2 \| 5d \rangle = 9.76(29)$ matrix element that was extracted from the K splittings of the bound $6snl$ states in Ref. [13] corresponds to the lifetime $\tau_{5d3/2} = 170(10)$ s, which is a factor of 2 longer than all other values. We discuss the lifetimes of the $5d_{3/2}$ and $5d_{5/2}$ levels in the next section.

V. LIFETIMES

The lifetime of a state a is calculated as $\tau_a = (\Sigma_{b\leqslant a} A_{ab})^{-1}$. The $E1$, $E2$, and $M1$ transition rates A_{ab} are given by [51]

$$A_{ab}^{E1} = \frac{2.02613 \times 10^{18}}{\lambda^3} \frac{S_{E1}}{2j_a + 1} \ \mathrm{s}^{-1}, \tag{8}$$

$$A_{ab}^{E2} = \frac{1.11995 \times 10^{18}}{\lambda^5} \frac{S_{E2}}{2j_a + 1} \ \mathrm{s}^{-1}, \tag{9}$$

$$A_{ab}^{M1} = \frac{2.69735 \times 10^{13}}{\lambda^3} \frac{S_{M1}}{2j_a + 1} \ \mathrm{s}^{-1}, \tag{10}$$

respectively, where λ is the wavelength of the transition in Å and S is the line strength. In this work, we calculated the lifetimes of the $6p_{1/2}$, $6p_{3/2}$, $5d_{3/2}$, and $5d_{5/2}$ levels in Ba$^+$. The results are compared with experimental and other theoretical values in Table V. Since the $6p$ levels are above $5d$ levels in Ba$^+$, we also needed to calculate the SD all-order reduced matrix elements for the $6p$-$5d$ $E1$ transitions, and our results (in atomic units) are $d(6p_{1/2}$-$5d_{3/2}) = 3.034$, $d(6p_{3/2}$-$5d_{3/2}) = 1.325$, and $d(6p_{3/2}$-$5d_{5/2}) = 4.080$. These values include contributions from the higher partial waves (0.6%) and 0.1%–0.2% Breit correction. The correlation corrections to these transitions are similar to the ones for the $6s$-$6p_j$ transitions. Therefore, similar (on the order of 0.5%) accuracy is expected for these matrix elements. The $6s$-$6p_j$ transitions contribute about 73% to the respective $\Sigma_{b\leqslant a} A_{ab}$ totals for the $6p_j$ lifetimes. Based on our evaluation of the uncertainty in these matrix elements discussed in Sec. III, we expect present $6p$ lifetime values to be accurate to about 1%. Our results are in excellent agreement with other recent theoretical [48,49] and experimental [24–26] values. The calculation of Refs. [46,47] is a third-order MBPT calculation that omits higher-order corrections included in the present calculation; slightly different values are expected.

Only one transition contributes to the $5d_{3/2}$ lifetime: the $6s$-$5p_{3/2}$ $E2$ transition (the contribution of the $6s$-$5d_{3/2}$ $M1$ transition is negligible). In the case of the $5d_{5/2}$ lifetime, the $M1$ $5d_{5/2}$-$5d_{3/2}$ transition has to be included as pointed out in [47,48,50]. Our SD all-order value for this transition (in a.u.) is 1.5493. The correlation correction contribution is very small, and the lowest order gives essentially the same value, 1.5489. The $M1$ transition contributes 17% to the $\Sigma_{b\leqslant a} A_{ab}$ total for the $5d_{5/2}$ level.

TABLE V. Lifetimes of the $6p_j$ and $5d_j$ states in Ba$^+$; comparison with experiment and other theory. The lifetimes of the $6p_j$ states are given in ns, and the lifetimes of the $5d_j$ states are given in s.

	$\tau_{6p_{1/2}}$ (ns)	$\tau_{6p_{3/2}}$ (ns)	$\tau_{5d_{3/2}}$ (s)	$\tau_{5d_{5/2}}$ (s)
Present	7.83	6.27	81.5(1.2)	30.3(4)
Theory [46,47]	7.99	6.39	83.7	30.8
Theory [48]	7.89	6.30	81.5	30.3
Theory [49]	7.92	6.31	81.4	36.5
Theory [50]			80.1(7)	29.9(3)
Theory [15]			82.0	31.6
Expt. [24]		6.312(16)		
Expt. [25]	7.92(8)			
Expt. [26]	7.90(10)	6.32(10)		
Expt. [17]			17.5(4)	
Expt. [18]			48(6)	
Expt. [19]			79.8(4.6)	
Expt. [18]			89.4(15.6)	32.0(4.6)
Expt. [20]				47.0(16)
Expt. [21]				32.0(5)
Expt. [22]				34.5(3.5)

We compare our final results for the $5d_{3/2}$ and $5d_{5/2}$ lifetimes with experimental [15,17–22] and other theoretical [15,46–50] values in Table V. We note that the calculation [49] omitted the $5d_{5/2}$-$5d_{3/2}$ $M1$ contribution to the $5d_{5/2}$ lifetime, leading to a higher value, as noted in later work [50]. We note that the fact that we quote slightly larger theoretical errors than Ref. [50] does not imply that our calculation is less precise than that of Ref. [50] (or earlier work [49]). Since it is a theoretical uncertainty, it is an estimate of the omitted (unknown) corrections. To compare the accuracy of our calculation and those of Refs. [49,50], we compare the effects that are included in our work and that of [49,50]. Our method includes some triple excitations and estimates of higher-order effects that were not included in those coupled-cluster calculations. We omit some nonlinear terms present in calculation of [49,50]. We also include contributions from higher partial waves and Breit interaction. The latter contribution was omitted in all previous works. We have also made an assumption that the uncertainties in all other terms do not exceed the uncertainty in the dominant terms (which can be reliably estimated). The uncertainty in Ref. [50] only accounts for the dominant term uncertainty. Our results are in agreement with other theoretical calculations and most recent values from [15] measured in a beam-laser experiment performed at the ion storage ring CRYRING, as well as experimental values from [19,21,22].

VI. CONCLUSION

In conclusion, we carried out the relativistic all-order calculations of Ba$^+$ $6s$-np_j $(n=6-9)$, $6p_{1/2}$-$5d_{3/2}$, $6p_{3/2}$-$5d_{5/2}$, and $6p_{3/2}$-$5d_{5/2}$ electric-dipole matrix elements; $6s$-$5d_{3/2}$,

$6s$-$5d_{5/2}$, $6s$-$6d_{3/2}$, $6s$-$6d_{5/2}$, $6s$-$7d_{3/2}$, and $6s$-$7d_{5/2}$ electric-quadrupole matrix elements; and $5d_{5/2}$-$5d_{3/2}$ magnetic-dipole matrix element. These values are used to evaluate lifetimes of the $6p_{1/2}$, $6p_{3/2}$, $5d_{3/2}$, and $5d_{5/2}$ levels as well as dipole and quadrupole ground-state polarizabilities. Extensive comparison with other theoretical and experimental values is carried out. The present values of the dipole polarizability and $6p_j$ lifetimes are in excellent agreement with experimental values. We estimated the uncertainty of our theoretical values for these properties to be on the order of 1%. Our recommended value of the quadrupole ground-state polarizability, $\alpha_{E2}=4182(34)a_0^5$, is in agreement with the most recent experimental work [14]. Because of the discrepancies in the experimental values of the $6s$-$5d$ matrix elements and quadrupole polarizability, we estimated some omitted corrections and conducted extensive analysis of the uncertainties in our calculations of these properties to establish the accuracy of our results. This uncertainty analysis is essential for a comprehensive comparison of the present values with different experiments. Our recommended values for the $5d_j$ lifetimes $\tau_{5d_{3/2}}=81.5(1.2)$ s and $\tau_{5d_{5/2}}=30.3(4)$ s are in agreement with other theoretical calculations and most recent values from [15] measured in a beam-laser experiment performed at the ion storage ring CRYRING, as well as experimental values from [19,21,22].

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