Ba+ quadrupole polarizabilities: Theory versus experiment

B. K. Sahoo[*]

Theoretical Physics Division, Physical Research Laboratory, Ahmedabad-380009, India

B. P. Das

Theoretical Astrophysics Group, Indian Institute of Astrophysics, Bangalore-560034, India

Three different measurements have been reported for the ground-state quadrupole polarizability in singly ionized barium (Ba+) which disagree with each other. Our calculation of this quantity using the relativistic coupled-cluster method disagrees with two of the experimental values and is within the error bars of the other. We discuss the issues related to the accuracy of our calculations and emphasize the need for further experiments to measure the quadrupole polarizability for this state and/or the $5D$ states.

I. INTRODUCTION

High-precision studies of polarizabilities of atomic systems are of interest in a number of different problems in physics [1–8]. A wide variety of methods have been used to calculate the polarizabilities of these systems [7–9]. Calculations of polarizabilities depend on the matrix elements between different atomic states and the excitation energies between them. Therefore, the accuracies of these calculations depend on the uncertainties in both quantities. In general, it is very challenging to minimize these uncertainties. However, in a sum-over-states approach [10,11], the major uncertainties in evaluating the polarizabilities can be reduced by using the experimental energies. Furthermore, the accuracies of the matrix elements can also be improved by matching the results of the lifetimes and branching ratios of atomic states from sophisticated many-body calculations and high-precision measurements. For cases where the experimental results are not sufficiently accurate or if all the available measured results are not in agreement, it is not possible to test the accuracies of the *ab initio* results. It might be useful in such situations to employ the sum-over-states approach to evaluate the polarizabilities.

For the ground-state quadrupole polarizability in Ba+, the available experimental results [12–14] are not in agreement with each other or with the calculations that are reported in this work. Investigation of various properties using relativistic many-body methods for high-precision studies in this ion is useful in the context of a proposed parity-nonconservation experiment [15,16], searching for the nuclear anapole moment [17], estimation of the uncertainties for a proposed optical clock [18], determination of the nuclear octuple moment [19], etc. In this paper, we report the results of our calculations of the matrix elements between different atomic states of Ba+ using the relativistic coupled-cluster (RCC) method. We also intend to test the accuracies of some of the important matrix elements by using them to estimate the lifetimes of the $5D$ states and comparing with their corresponding measurements.

Before presenting our results, we define the quadrupole polarizability for a general atomic state in the following section.

We give a brief description of the method of calculations of the wave functions and the matrix elements in Sec. III and then present our calculated results and discussions in Sec. IV, after which we make our concluding remarks.

II. THEORY

The potential energy of an atom in the presence of a static electric field is given by (for example, see [7])

$$V(r) = -p_i \mathcal{E}_i - \frac{1}{6} Q_{ij} \partial_i E_j + \cdots = e \sum_i \mathcal{E}_i r^i P_i(\cos\theta), \quad (1)$$

where p_i and Q_{ij} are the components of the electric dipole and quadrupole operators, respectively, and $\mathcal{E}_i$ and $P_i(\cos\theta)$ are the applied electric field and Legendre polynomial, respectively, with component i. The quadrupole polarizability of an atomic state $|\Psi_n\rangle$ is related to the energy shift by the equation [20,21]

$$\delta E_n(\mathcal{E}) = -\frac{1}{8}\alpha_2^n \mathcal{E}^2, \quad (2)$$

where α_2^n is known as the quadrupole polarizability of the state $|\Psi_n\rangle$, which is defined as

$$\alpha_2^n = -2 \sum_{m\neq n} \frac{|\langle \Psi_n | Q | \Psi_m \rangle|^2}{E_m - E_n}, \quad (3)$$

and $Q = \sum q$ is the total electric quadrupole operator whose single-particle reduced matrix element is given by

$$\langle \kappa_f || q || \kappa_i \rangle = \langle \kappa_f || C^{(2)} || \kappa_i \rangle$$
$$\times \int_0^\infty dr\, r^2 \left[P_f(r) P_i(r) + Q_f(r) Q_i(r) \right], \quad (4)$$

where $P(r)$ and $Q(r)$ represent the radial parts of the large and small components of the single-particle Dirac orbitals, respectively. The reduced Racah coefficients are given by

$$\langle \kappa_f || C^{(k)} || \kappa_i \rangle = (-1)^{j_f + 1/2} \sqrt{(2j_f+1)(2j_i+1)}$$
$$\times \begin{pmatrix} j_f & k & j_i \\ 1/2 & 0 & -1/2 \end{pmatrix} \pi(l_{\kappa_f}, k, l_{\kappa_i}), \quad (5)$$

[*]bijaya@prl.res.in

with

$$\pi(l,m,l') = \begin{cases} 1 & \text{for} \quad l+m+l' = \text{even}, \\ 0 & \text{otherwise}. \end{cases} \tag{6}$$

III. METHOD OF CALCULATIONS

In the present study, the atomic states of Ba$^+$ that we have considered have closed-shell cores and a valence electron v. We adopt a two-step procedure to calculate the wave functions for these states in the Fock space representation: First, the Dirac-Fock (DF) wave function for the common closed-shell core $[5p^6]$ (denoted by $|\Phi_0\rangle$) is calculated. In the next step, we append a corresponding valence orbital v to obtain the new DF wave function for the new configuration (denoted by $|\Phi_v\rangle = a_v^\dagger|\Phi_0\rangle$). The atomic state for the new configuration is expressed using the RCC ansatz as

$$|\Psi_v\rangle = e^T\{1 + S_v\}|\Phi_v\rangle, \tag{7}$$

where T and S_v are the excitation operators that take into account correlation effects arising from the core and valence electrons, respectively. We consider only the singly and doubly excited configurations from the DF wave functions by approximating the T and S_v operators as

$$T = T_1 + T_2, \quad S_v = S_{1v} + S_{2v}. \tag{8}$$

The above approximation is known as the coupled-cluster singles and doubles (CCSD) method. Also, we construct triple-excitation configurations from $|\Phi_v\rangle$ perturbatively in the spirit of the CCSD with partially triple excitations [CCSD(T)] approach [9,16,22–26].

The amplitudes for both the T and S_v operators are determined using the following equations:

$$\left\langle\Phi_0^K\left|\{\widehat{H_N e^T}\}\right|\Phi_0\right\rangle = \delta_{0,K}\Delta E_{corr} \tag{9}$$

and

$$\begin{aligned}\left\langle\Phi_v^K\left|\{\widehat{H_N e^T}\}\{1 + S_v\}\right|\Phi_v\right\rangle &= \left\langle\Phi_v^K\left|1 + S_v\right|\Phi_v\right\rangle \\ &\times \left\langle\Phi_v\left|\{\widehat{H_N e^T}\}\{1 + S_v\}\right|\Phi_v\right\rangle \\ &= [\delta_{v,K} + \langle\Phi_v^K|S_v|\Phi_v\rangle]\Delta E_v^{att},\end{aligned} \tag{10}$$

respectively. Here $K = 1,2,\dots$ represents the singly, doubly, etc., excited configurations with respect to their corresponding reference states, $\widehat{H_N e^T}$ denotes the connected terms of the normal-order Dirac-Coulomb (DC) Hamiltonian H_N with the T operators, and ΔE_{corr} and ΔE_v^{att} are the correlation energies for the closed-core and attachment energies of the valence electron v, respectively.

We evaluate the scalar polarizabilities by expressing them as the sum of three contributions:

$$\alpha_2^v = \alpha_2^v(c) + \alpha_2^v(cv) + \alpha_2^v(v), \tag{11}$$

where $\alpha_q^v(c)$ takes into account the contributions of the core orbitals and $\alpha_q^v(cv)$ and $\alpha_q^v(v)$ are the core-valence and valence contributions, respectively. In terms of the reduced matrix

elements, the expressions of these parts are given by

$$\alpha_2^v(c) = -\frac{2}{5}\sum_{c,p}\frac{|\langle J_p||Q||J_c\rangle|^2}{E_c - E_p}, \tag{12}$$

$$\alpha_2^v(cv) = -\frac{2}{5(2J_v + 1)}\sum_c\frac{|\langle J_v||Q||J_c\rangle|^2}{E_c - E_v}, \tag{13}$$

and

$$\alpha_2^v(v) = -\frac{2}{5(2J_v + 1)}\sum_{m\neq v}\frac{|\langle J_v||Q||J_m\rangle|^2}{E_m - E_v}, \tag{14}$$

where $\langle J_p||Q||J_c\rangle$ are the reduced matrix elements between the atomic states with angular momenta J_p and J_c. $\alpha_2^v(c)$ and $\alpha_2^v(cv)$ have been calculated using the third-order many-body perturbation theory [MBPT(3) method] from the expression

$$\alpha_2^n = \langle\Psi_n|Q|\Psi_n^{(1)}\rangle, \tag{15}$$

where $|\Psi_n^{(1)}\rangle$ is like a first-order perturbed wave function and it is obtained by solving the following inhomogeneous equation:

$$(H - E_n)\big|\Psi_n^{(1)}\big\rangle = \big(E_n^{(1)} - Q\big)|\Psi_n\rangle, \tag{16}$$

with $E_n^{(1)} = \langle\Psi_n|Q|\Psi_n\rangle$, which is similar to the first-order perturbation equation. It should be noted that unlike for the dipole operator [9,27], $E_n^{(1)}$ results are finite for the quadrupole operator and correspond to the quadrupole moments of the respective states $|\Psi_n\rangle$.

Contributions from $\alpha_2^v(v)$ were determined by calculating important intermediate states explicitly using the CCSD(T) method. The reduced matrix elements between different states were computed using the following expression:

$$\langle J_f||Q||J_i\rangle = \frac{\langle J_f||\{1 + S_f^\dagger\}\overline{Q}\{1 + S_i\}||J_i\rangle}{\sqrt{\mathcal{N}_f\mathcal{N}_i}}, \tag{17}$$

where $\overline{Q} = e^{T^\dagger}Q e^T$ and $\mathcal{N}_v = \langle\Phi_v|e^{T^\dagger}e^T + S_v^\dagger e^{T^\dagger}e^T S_v|\Phi_v\rangle$ involve two nontruncating series in the above expression. The details of the calculations of these terms are discussed elsewhere [9,16,22,23].

IV. RESULTS AND DISCUSSIONS

Below we present the quadrupole polarizabilities for the ground and the first two excited D states in Ba$^+$. We have used the experimental energies from the National Institute of Science and Technology (NIST) database [28] in the sum-over-states approach to evaluate the major contributions to the quadrupole polarizabilities, which are the valence correlation effects. The main purpose of doing this is, as stated in Sec. I, to minimize the uncertainties in the calculated results so that we shall be able to compare them meaningfully with the available experimental results. Contributions from the higher excited states that cannot be accounted for by the valence correlation in the sum-over-states approach are evaluated using the MBPT(3) method since their contributions are typically smaller. The uncertainties in these results are estimated by scaling results obtained using this method with the CCSD(T) calculations.

In Table I, we present the results of our quadrupole polarizability calculations of the ground state of Ba$^+$. It is clear

TABLE I. The $E2$ matrix elements and the ground-state quadrupole polarizability in Ba$^+$ (in a.u.) Results are given up to significant digits. Possible uncertainties in the results are given in parentheses.

Transition		Amplitude	α^{E2}
$\alpha_2^{6s}(v)$			
$6s_{1/2} \rightarrow$	$5d_{3/2}$	12.76(5)	1466(11)
	$6d_{3/2}$	16.58(12)	263(4)
	$7d_{3/2}$	5.727(7)	24.07(6)
	$8d_{3/2}$	4.036(5)	10.72(3)
	$5d_{5/2}$	15.99(8)	1978(20)
	$6d_{5/2}$	19.99(20)	380(8)
	$7d_{5/2}$	7.024(9)	36.1(1)
	$8d_{5/2}$	5.022(5)	16.59(3)
$\alpha_2^{6s}(c)$			46(2)
$\alpha_2^{6s}(cv)$			$-0.001(0)$
$\alpha_2^{6s}(\text{tail})$			50(10)
$\alpha_2^{6s}(\text{total})$			4270(27)
Experiment [12]			2050(100)
Experiment [13]			2462(361)
Experiment [14]			4420(250)
Theory [29]			4091.5
Theory [30]			4182(34)
Theory [31]			4821

from Table I that the dominant contributions (approximately 80%) come from the $5D$ states. Thus the accuracy of the ground-state quadrupole polarizability calculation depends primarily on the accurate determination of the $E2$ matrix elements of the $6S_{1/2} \rightarrow 5D_{3/2}$ and $6S_{1/2} \rightarrow 5D_{5/2}$ transitions. These matrix elements were also calculated by us earlier using the same CCSD(T) method but with different basis functions [16,32], and all the results are in good agreement. There are also other calculations available for these matrix elements using different variants of the RCC methods and basis functions [29,30,33,34]; all the calculated results seem to be in reasonable agreement with each other. Moreover, the accuracies of these matrix elements can be verified by using them to estimate the lifetimes of the $5D$ states and comparing them with the measurements as stated in the Introduction. Possible transition channels from the $5D_{3/2}$ state to the ground state are due to the $M1$ and $E2$ multipoles. As we have shown in our earlier work [32], the $M1$ transition probability is very small in this case, and its contribution to the lifetime of the $5D_{3/2}$ state is negligible. Therefore, neglecting the $M1$ contribution and using our calculated $E2$ matrix element of the $6S_{1/2} \rightarrow 5D_{3/2}$ transition, we obtain the lifetime of the $5D_{3/2}$ state as 79.8(6) s. The measured values are 79.8(4.6) s [35] and 89.4(15.6) s [36]. Our result is in good agreement with the first experimental result and is within the error bars of the second result. To calculate the lifetime of the $5D_{5/2}$ state, it is necessary to take into account all the $M1$ and $E2$ transition probabilities from this state to the $6S_{1/2}$ and $5D_{3/2}$ states. We have also shown in Ref. [32] that only the $E2$ and $M1$ transition probabilities of the $5D_{5/2} \rightarrow 6S_{1/2}$ and $5D_{5/2} \rightarrow 5D_{3/2}$ transitions, respectively, are significant in the determination the lifetime of the $5D_{5/2}$ state. In the present

work, we have calculated the above $M1$ transition amplitude to be 1.544(1) a.u., which is in agreement with the result reported in Ref. [32]. Using this value we find the lifetime of the $5D_{5/2}$ state to be 29.8(3) s with an 84% branching ratio to the $5D_{5/2} \rightarrow 6S_{1/2}$ transition. This is also in agreement with the experimental results, which are reported as 34.5(3.5) s [35], 31.6(4.6) s [36], and 32(2) s [37]. We have used the experimental wavelengths to determine the lifetimes of the $5D$ states in order to verify the accuracies of the calculated $E2$ matrix elements. This analysis suggests that our calculated $E2$ matrix elements are accurate to within a few percent, and therefore they can be considered for high-precision studies of the quadrupole polarizabilities of Ba$^+$.

The next significant contributions to the ground-state quadrupole polarizability come from the $6D$ states. It is difficult to estimate the accuracies of the $E2$ matrix elements of the $6S_{1/2} \rightarrow 6D_{3/2}$ and $6S_{1/2} \rightarrow 6D_{5/2}$ transitions from the measured lifetimes of the $6D$ states because of their negligible roles in the theoretical determination of these lifetimes. Also, the contributions from the higher excited states (tail) and core correlations to the final result of the quadrupole polarizability of the ground state in Ba$^+$ are non-negligible. A suitable method to test the validity of all these contributions is to compare the final calculated result with the available measurements. The reported experimental results for the ground-state quadrupole polarizability of Ba$^+$ are 2050(100) a.u. [12], 2462(361) a.u. [13], and 4420(250) a.u. [14]. The first two results agree with each other, but they are completely in disagreement with the latest result. All the reported experimental results have relatively large error bars. Two of these experimental results are just half of our calculated result. In such a situation, it will not be possible to test the accuracies of the many-body methods that have been used to perform the calculations as well as those that are are likely to be developed in the *ab initio* framework to calculate these quantities. Also, the above experimental techniques could single out the contributions from the $5D$ states to the ground-state quadrupole polarizability, and their values have been reported as 1562(93) a.u. [38], 2050(100) a.u. [12], 1828(88) a.u. [13], and 1524(8) a.u. [14]. On the other hand, using the $E2$ matrix elements of the $6S_{1/2} \rightarrow 5D_{3/2}$ and $6S_{1/2} \rightarrow 5D_{5/2}$ transitions, whose accuracies have been discussed above, we obtain the combined contributions of the $5D$ states to the ground-state quadrupole polarizability in Ba$^+$ as 3444(23) a.u. with individual contributions as 1466(11) and 1978(20) a.u. from the $5D_{3/2}$ and $5D_{5/2}$ states, respectively. It therefore appears that the extracted values of these contributions of the $5D$ states to the ground-state quadrupole polarizabilities in the above experimental analysis may not be the contributions from both the $5D$ states, but rather from either the $5D_{3/2}$ or the $5D_{5/2}$ state individually in the different experiments.

To the best of our knowledge, three calculations of the ground-state quadrupole polarizability in Ba$^+$ have been carried out, yielding 4091.5 a.u. [29], 4182(34) a.u. [30], and 4821 a.u. [31]. All these results are in agreement with our calculation; the first two are obtained by the linearized all order singles, doubles and partial triples (SDpT) RCC method, in contrast to our nonlinear approach. The two results differ from each other by about 2%, even though the same

SDpT method was employed in the two cases, and they are at the lower limit of the latest experimental result, 4420(250) a.u. Their calculated results are slightly lower than our result of 4270(27) a.u., and also, they do not overlap within their respective predicted uncertainties. The main reason for the difference between the SDpT method and our results is that the calculated values of the $E2$ matrix elements of the $6S_{1/2} \rightarrow 5D_{3/2}$ and $6S_{1/2} \rightarrow 5D_{5/2}$ transitions for the two cases are different. It is necessary to emphasize that the precision of the theoretical determination of the lifetimes of the $5D$ states depends critically on the accuracies of these matrix elements [29,30,32–34]. A precise measurement of the quadrupole polarizability of the ground state in Ba$^+$ could test the accuracies of these $E2$ matrix elements that are calculated by different many-body methods. A third calculation of the ground-state quadrupole polarizability has reported a result of 4821 a.u. [31] using rather simple wave functions based on the asymptotic behavior and the binding energy of the valence electron, in contrast to other calculations which are based on all order perturbative methods in the RCC framework. Nevertheless, all the calculated results suggest that the ground-state quadrupole polarizability in Ba$^+$ is approximately 4200 a.u., and all the $E2$ matrix elements of the $6S_{1/2} \rightarrow 5D_{3/2}$ and $6S_{1/2} \rightarrow 5D_{5/2}$ transitions are in reasonable agreement with each other. In view of the discrepancies between the calculated and experimental results as well as among the individual values of the latter, it would indeed be desirable to perform precise measurements of the quadrupole polarizability of the ground state of Ba$^+$.

An alternative approach to resolve this problem would be to measure the scalar quadrupole polarizabilities of the $5D$ states with high precision and attempt to identify the individual contribution of the $6S$ state, perhaps in a manner similar to the method used in obtaining the contributions of the $5D$ states to the quadrupole polarizability of the ground state. If that

TABLE II. The quadrupole polarizabilitiy of the $5D_{3/2}$ state of Ba$^+$ (in a.u.). Uncertainties are given in parenthesis.

Transition		Amplitude	α^{E2}
$\alpha_2^{5d3/2}(v)$			
$5d_{3/2} \rightarrow$	$6s_{1/2}$	12.76(5)	−733(6)
	$7s_{1/2}$	4.882(5)	13.96(3)
	$8s_{1/2}$	1.558(2)	1.0(2)
	$9s_{1/2}$	1.360(1)	0.668(2)
	$6d_{3/2}$	8.36(4)	37.3(5)
	$7d_{3/2}$	3.021(3)	3.65(1)
	$8d_{3/2}$	2.183(2)	1.69(1)
	$5d_{5/2}$	6.83(2)	1278(7)
	$6d_{5/2}$	5.349(2)	15.21(1)
	$7d_{5/2}$	1.978(2)	1.56(1)
	$8d_{5/2}$	1.431(2)	0.727(3)
	$5g_{7/2}$	8.45(8)	26.9(5)
	$6g_{7/2}$	8.63(9)	25.7(5)
$\alpha_2^{5d3/2}(c)$			46(2)
$\alpha_2^{5d3/2}(cv)$			−0.49(3)
$\alpha_2^{5d3/2}(\text{tail})$			116(30)
$\alpha_2^{5d3/2}(\text{total})$			835(32)

TABLE III. The quadrupole polarizability of the $5D_{5/2}$ state in Ba$^+$ (in a.u.). Estimated uncertainties for the results are given in parentheses.

Transition		Amplitude	α^{E2}
$\alpha_2^{5d5/2}(v)$			
$5d_{5/2} \rightarrow$	$6s_{1/2}$	15.99(8)	−659(6)
	$7s_{1/2}$	6.409(7)	16.39(3)
	$8s_{1/2}$	1.992(2)	1.109(3)
	$9s_{1/2}$	1.723(2)	0.724(2)
	$5d_{3/2}$	6.83(2)	−852(5)
	$6d_{3/2}$	5.754(5)	12.03(3)
	$7d_{3/2}$	2.030(2)	1.114(2)
	$8d_{3/2}$	1.457(2)	0.509(1)
	$6d_{5/2}$	11.26(2)	45.8(2)
	$7d_{5/2}$	4.029(3)	4.38(1)
	$8d_{5/2}$	2.925(3)	2.05(1)
	$5g_{7/2}$	2.96(5)	2.24(8)
	$6g_{7/2}$	3.00(6)	2.1(1)
	$5g_{9/2}$	10.48(8)	28.0(4)
	$6g_{9/2}$	10.62(9)	26.3(5)
$\alpha_2^{5d5/2}(c)$			46(2)
$\alpha_2^{5d5/2}(cv)$			−0.48(3)
$\alpha_2^{5d5/2}(\text{tail})$			121(35)
$\alpha_2^{5d5/2}(\text{total})$			−1201(36)

could be achieved, then it would be possible to extract the $E2$ matrix elements of the $6S_{1/2} \rightarrow 5D_{3/2}$ and $6S_{1/2} \rightarrow 5D_{5/2}$ transitions. The advantages of measuring the scalar quadrupole polarizabilities of the $5D$ states in addition to the ground state could be of twofold: (i) The core-correlation effects, which are one of the significant contributions and are the same for the states that we have considered, can be ignored in the estimation of the accuracies of the $E2$ matrix elements. (ii) By combining the quadrupole polarizabilities with the lifetimes of the $5D$ states, it would be possible to extract the $E2$ matrix elements of the above two transitions. For this purpose, we also calculate the quadrupole polarizabilities of the $5D$ states.

In Table II, we present the scalar quadrupole polarizability of the $5D_{3/2}$ state. The largest contribution to this quantity comes from its fine-structure partner followed by the $6S$ state, but with opposite sign, resulting in a strong cancellation between them. The other significant contribution is from the higher excited g states (given as tail), which are not taken into account explicitly in the sum-over-states approach. Also, we find the trend of the contributions to the $5D_{5/2}$-state scalar quadrupole polarizability, given in Table III, is similar to those for the $5D_{3/2}$ state, but the contributions from this state and the $6S$ state have same sign. The contributions from the high-lying g states are significant, and it would be appropriate to use a method that implicitly takes all possible intermediate states into account, perhaps similar to Ref. [39], for an accurate evaluation of the scalar quadrupole polarizabilities of the $5D$ states. Nonetheless, the present study captures several classes of important correlation effects, and it will be useful in guiding experiments on quadrupole polarizabilities in Ba$^+$.

V. CONCLUSION

We have carried out a detailed analysis of the calculated and experimental results of the reported ground-state quadrupole polarizability in Ba$^+$ and highlighted the disagreement between the different studies. The reported experimental results are not reliable enough to test the validity of the calculated results. On the basis of different physical considerations, we propose new theoretical and experimental studies of the quadrupole polarizabilities of the $5D$ states to test the accuracies of the $E2$ matrix elements between the ground state and the $5D$ states and the quadrupole polarizability of the ground state. We have also presented the results of our calculations for the quadrupole polarizability of the $5D$ states in Ba$^+$ using the $E2$ transition amplitudes obtained from our calculations.

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