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Relativistic coupled-cluster theory of quadrupole moments and hyperfine structure constants of $5d$ states in Ba$^+$

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The narrow optical frequency resonances of the $6s\,^2S_{1/2} \rightarrow 5d\,^2D_{3/2}$ and $6s\,^2S_{1/2} \rightarrow 5d\,^2D_{5/2}$ forbidden transitions in Ba$^+$ have been proposed as suitable frequencies for a new optical frequency standard. The major sources of errors in measurements of singly ionized systems are due to the quadratic Zeeman and electric quadrupole shifts. We report here the most accurate calculations to date for the hyperfine structure constants and electric-quadrupole moments of the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states in Ba$^+$, which determine the quadratic Zeeman and electric quadrupole shifts. Relativistic coupled-cluster theory has been employed to calculate these quantities and large electron correlation effects are observed. It is also shown that for the high accuracy calculation of the $5d\,^2D_{5/2}$ state in Ba$^+$, all order core polarization effects play a significant role.

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During the last two decades searches have been carried out to find suitable single ions that could be used for the optical frequency standards [1–4]. A very narrow optical atomic resonance of the forbidden transitions with quality factor (Q) larger than 10^{15} can be suitable for the clock transitions to make precise and stable oscillators. Singly ionized atomic systems are suitable for high-resolution optical spectroscopy because of their stabilization in a perturbation free environment. They serve as high quality references largely free from shifts and perturbations which mainly come from the Doppler effects. The most important proposed candidates are In$^+$ [5], Hg$^+$ [2,6], Yb$^+$ [3], Sr$^+$ [4,7], and Ca$^+$ [8]. Single ionized barium (Ba$^+$) has been trapped [9,10] and its energy levels as opposed to other ions are easily accessible by lasers (see Fig. 1). Hence, many sophisticated measurements have been carried out on Zeeman resonances [9], light shifts [10], Stark shifts [11], etc., in this ion. A sensitive experiment on parity nonconservation (PNC) has also been proposed [12]. Lifetimes of metastable $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states are around 80 s [11] and 30 s [13], respectively. Due to its suitability to conduct high-precision measurements, it has recently been proposed to measure the atomic clock frequencies of the $6s\,^2S_{1/2} \rightarrow 5d\,^2D_{3/2}$ and $6s\,^2S_{1/2} \rightarrow 5d\,^2D_{5/2}$ forbidden transitions [14,15]. The main sources of systematic errors to measure these frequencies are due to linear Zeeman shift, quadratic Zeeman shift, second-order Stark shift, and electric quadrupole shift in the external electromagnetic field [16]. The largest source of errors arises from the electric quadrupole shift. Precise values of electric quadrupole moments (Θ) in atomic systems can be used to determine this shift accurately. Sherman *et al.* mentioned that their apparatus might also be used to measure the electric quadrupole moment of the $5d\,^2D_{3/2}$ state in Ba$^+$ [10], but the experiment is in a build-up phase [17]. Indeed, earlier experiments to measure Θ in ^{88}Sr$^+$ [7], ^{199}Hg$^+$ [18], and ^{171}Yb$^+$ [19] show that these instruments are complicated and it is difficult to measure this quantity precisely [7]. As has been mentioned by Itano, the calculated results are useful, even though they are not very precise [16,20]. Accurate studies of hyperfine structure constants can also be used to determine the quadratic Zeeman effect [16]. These results are also used to determine the accuracy of the PNC matrix elements in Ba$^+$ [21]. Especially, an accurate calculation of the $5d\,^2D_{5/2}$ state is very useful for the proposed spin-dependent PNC study in this system [22]. In principle, *ab initio* calculations of the above physical quantities can also test the many-body theory. A powerful many-body theory that is capable of incorporating the correct scale of electron correlation effects in heavy systems can be used for precise determination of atomic properties.

We employ here the relativistic coupled-cluster (RCC) theory, referred to as a universal many-body theory [23] and widely applied for precision studies in a broad range of physics [24,25], to calculate hyperfine structure constants and Θ of the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states in Ba$^+$. In contrast to theories like configuration interaction (CI) method, RCC theory has size-consistent and size-extensivity behavior. It is an all order theory and capable enough to consider relativistic and correlation effects rigorously. This theory has been successfully applied to high precision studies of some subtle effects like PNC [26] and electric dipole moment (EDM) [27] in atomic systems.

In an atomic state $|\Psi(\gamma, JM)\rangle$ with total angular momentum J and its maximum value of the magnetic quantum number $M=J, \Theta$ is given by [16]

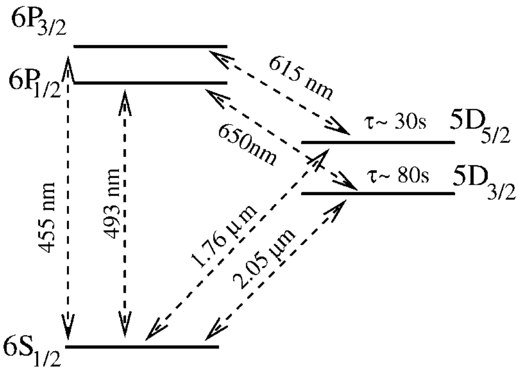

FIG. 1. Energy levels of Ba$^+$ showing the different transitions between $6s$, $6p$, and $5d$ states.

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$$\Theta = \langle \Psi(\gamma, JJ | \Theta_{zz}^{(2)} | \Psi(\gamma, JJ) \rangle, \tag{1}$$

with $\Theta_{zz}^{(2)}$ is the z component of a second rank electric quadrupole operator for the atom and is given by

$$\Theta_{zz}^{(2)} = \sum_m \sum_{q=-2}^{2} \theta_{m,q}^{(2)} = -\frac{e}{2}\sum_m (3z_m^2 - r_m^2). \tag{2}$$

Here z_m and r_m are the coordinate and radial distance of the mth electron. The reduced matrix element of this operator, in terms of the single particle orbitals in the lowest order approximation, is

$$\langle j_f \| \theta_{m,q}^{(2)} \| j_i \rangle = \langle j_f \| C_q^{(2)} \| j_i \rangle \int dr_m r_m^2 (P_{\kappa_f} P_{\kappa_i} + Q_{\kappa_f} Q_{\kappa_i}), \tag{3}$$

where j's and κ's are the total and relativistic angular momenta of the single particle orbitals, the C_q^k represents Racah angular momentum operator for rank, k and P_{κ_i} and Q_{κ_i} represent the large and small components of the Dirac wave function.

In order to determine quadratic Zeeman shift, precise data on hyperfine structure constants are necessary [16]. Studies of hyperfine interactions in heavy systems also serve as a stringent test of atomic wave functions in the nuclear region. This can be used here to estimate the corrections of the wavefunctions from the deviation of the calculated results and the measured values so that quantities such as Θ, whose experimental results are not known in Ba$^+$, can be calculated within some uncertainties.

The magnetic dipole (A) and electric quadrupole (B) hyperfine structure constants are given by [28]

$$A = \mu_N g_I \frac{\langle J \| \mathbf{T}^{(1)} \| J \rangle}{\sqrt{J(J+1)(2J+1)}} \tag{4}$$

and

$$B = 2eQ \left\{ \frac{2J(2J-1)}{(2J+1)(2J+2)(2J+3)} \right\}^{1/2} \langle J \| \mathbf{T}^{(2)} \| J \rangle, \tag{5}$$

where Q is the nuclear quadrupole moment and $g_I = \left[\frac{\mu_I}{I}\right]$ is the nuclear g factor with the nuclear magnetic moment μ_I and spin I. In the above expression μ_N is the Bohr magneton and $\mathbf{T}^{(k)} = \Sigma_m \Sigma_{q=-k}^{k} t_{m,q}^{(k)}$ are the hyperfine interaction operators with $k=1$ for A and $k=2$ for B, which in terms of single particle orbitals are given by

$$\langle j_f \| t_{m,q}^{(1)} \| j_i \rangle = -(\kappa_f + \kappa_i) \langle j_f \| C_q^{(1)} \| j_i \rangle \int dr_m \frac{(P_{\kappa_f} Q_{\kappa_i} + Q_{\kappa_f} P_{\kappa_i})}{r_m^2}, \tag{6}$$

$$\langle j_f \| t_{m,q}^{(2)} \| j_i \rangle = -\langle j_f \| C_q^{(2)} \| j_i \rangle \int dr_m \frac{(P_{\kappa_f} P_{\kappa_i} + Q_{\kappa_f} Q_{\kappa_i})}{r_m^3}. \tag{7}$$

It is obvious from Eqs. (3), (6), and (7) that the radial dependencies are different for these properties. The RCC theory has been employed recently to carry out sub one percent calculations of quantities that depend simultaneously on different radial functions [26,27]. The wave functions using this theory for the single valence (v) system can be expressed as [29]

$$|\Psi_v\rangle = e^T \{1 + S_v\} |\Phi_v\rangle, \tag{8}$$

where $|\Phi_v\rangle$ is the open-shell Dirac-Fock (DF) wave function defined as $|\Phi_v\rangle = a_v^\dagger |\Phi_0\rangle$ and $|\Phi_0\rangle$ is the closed-shell DF wave function. T and S_v are the closed and open shell particle-hole excitation operators, respectively. The curly bracket represents the normal ordered form of the operators. We have developed fully nonlinearized singles and doubles with leading order triples relativistic coupled-cluster [CCSD(T)] codes and have been used to study accurate calculations in heavy systems [21,30].

We consider the frequency independent Dirac-Coulomb-Breit Hamiltonian given by

$$H = \sum_i^N \Lambda_i^+ [c\vec{\alpha}_i \cdot \vec{p}_i + (\beta_i - 1)c^2 + V_{\text{nuc}}(r_i)] \Lambda_i^+ + \sum_{i>j}^N \Lambda_i^+ \Lambda_j^+ \left\{ \frac{1}{r_{ij}} - \frac{1}{2}\frac{\vec{\alpha}_i \cdot \vec{\alpha}_j}{r_{ij}} - \frac{(\vec{\alpha}_i \cdot \vec{r}_{ij})(\vec{\alpha}_j \cdot \vec{r}_{ij})}{r_{ij}^3} \right\} \Lambda_j^+ \Lambda_i^+,$$

where the Λ^+s are the projection operators on to the positive-energy states of the Dirac Hamiltonian in the nuclear [$V_{\text{nuc}}(r_i)$] and DF [$U(r_i)$] potentials.

The expectation value of a Hermitian operator (O) in the single valence states using the RCC theory can be written as

$$\langle O \rangle = \frac{\langle \Psi_v | O | \Psi_v \rangle}{\langle \Psi_v | \Psi_v \rangle} = \frac{\langle \Phi_v | \{1 + S_v^\dagger\} \bar{O} \{1 + S_v\} | \Phi_v \rangle}{1 + N_v}, \tag{9}$$

where we define $\bar{O} = e^{T^\dagger} O e^T$ and $N_v = S_v^\dagger e^{T^\dagger} e^T S_v$. We calculate the above expression using the procedure followed in the earlier works [21,30]. Contributions from normalization factors are given by

$$(\text{Normalization factors}) = \langle \Psi_v | O | \Psi_v \rangle \left\{ \frac{1}{1 + N_v} - 1 \right\}. \tag{10}$$

The orbitals are constructed as linear combinations of finite basis functions of Gaussian type orbitals (GTOs) of the form [31]

$$F_{i,k}(r) = r^k e^{-\alpha_i r^2}, \tag{11}$$

where i represents the radial grid points and $k=0,1,\ldots,$ for $s,p,\ldots,$ type orbital symmetries, respectively. For the exponents, we use the even tempering condition as

$$\alpha_i = \alpha_0 \beta^{i-1}. \tag{12}$$

In our earlier work it was found that GTOs are able to produce accurate hyperfine structure constants in different sizes of systems [21,30]. The α_0 and β values considered in the present calculations are 0.00525 and 2.73, respectively, for all the symmetries. All core electrons are activated in the RCC calculations. We account for the finite size of the nucleus by considering the Fermi charge distribution given by

$$\rho(r) = \frac{\rho_0}{1 + e^{(r-c)/a}}, \tag{13}$$

where the fitted values of a and c parameters are used as the relations given by Parpia and Mohanty [32].

Since the atomic wave functions in ^{137}Ba$^+$ and ^{138}Ba$^+$ do not depend much on the nuclear parameters, the calculated

TABLE I. A comparison study of A and B in MHz of the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states using the Dirac-Fock, CCSD(T) and experimental results.

	$5d\,^2D_{3/2}$			$5d\,^2D_{5/2}$		
	DF	RCC	Exp. [36]	DF	RCC	Exp. [37]
A	128.34	190.89	189.8277(6)	51.61	−11.99	−12.028(11)
B	32.77	46.82	44.5417(16)	41.99	62.27	59.533(43)

quantities in these systems are almost same. Therefore, we discuss first the calculated results of A and B in ^{137}Ba$^+$ for the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states whose experimental data are available, then present the Θ results, which can be used for both the systems.

We have used g_I=0.6249 [33]. We obtain Q=0.246(1)b ($1b$=1 b=10^{-28} m^2) for ^{137}Ba by combining our calculated result B/Q=375.102 MHz b^{-1} of the $6p\,^2P_{3/2}$ state in ^{137}Ba$^+$ with the experimental result B=92.5(2) MHz [34]. The error has only been considered from the experimental result. This Q value is used latter to evaluate B for the $5d$ states. Very precise measured value of A for the $6s\,^2S_{1/2}$ state in ^{137}Ba$^+$ is available [35]. Again, due to the second rank operators in Eqs. (1) and (5) both Θ and B do not exist in the $6s\,^2S_{1/2}$ state. We are mainly interested in optical clock transitions, and therefore we calculate Θ, A, and B for the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states.

It has been shown that the d states have strong correlation effects unlike s and p states [30]. We have compared our DF and CCSD(T) results of A and B for the $5d$ states in Ba$^+$ with their corresponding experimental results in Table I. It shows that the DF values of A for the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states are far away from the experimental results. In fact, it is interesting to note that for the $5d\,^2D_{5/2}$ state the DF value of A and its experimental result have opposite signs. Correlation effects by the CCSD(T) method improve the A results from the DF values by 33% in the $5d\,^2D_{3/2}$ state and 530% in the $5d\,^2D_{5/2}$ state. Both the CCSD(T) results are well matched with their experimental results. From this comparison it can be concluded that the calculated wavefunctions can evaluate other properties accurately in these states. Therefore, Θ could be calculated using these wavefunctions for the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states in Ba$^+$.

We obtain Θ as $2.32a_0^2$ and $3.42a_0^2$ for the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states, respectively. We present contributions from different RCC terms in Table II for the A, B, and Θ. This table explains the importance of the correlation effects. The trend of the correlation effects for all the properties varies in both the states. It is clear from Tables I and II that although both B and Θ operators are second rank, the DF contributions to the former property is around 75%, while the latter has a higher value than the total obtained results in the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states. As explained in our earlier papers, $\bar{O}S_{1v}$ and $\bar{O}S_{2v}$, with their complex conjugate terms (c.c.), are the two most important correlation terms in the RCC theory [21,30]. The first term represents all order Brückner pair-correlation effects, whereas the second term gives all order core-polarization effects. Contributions from these two terms show different trends in A, B, and Θ. The core-polarization effects are stronger than the pair-correlation effects in A and B. The contributions from this effect in A calculation for the $5d\,^2D_{5/2}$ state is the largest among all other RCC terms and has opposite sign from the DF value. The cancellation of these two values determines the final result. In the case of Θ, for both the states, the pair-correlation effects seem to be more important.

TABLE II. Breakdown of the CCSD(T) contributions to A, B, and Θ of the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states in ^{137}Ba$^+$.

	$5d\,^2D_{3/2}$			$5d\,^2D_{5/2}$		
RCC terms	A	B	Θ	A	B	Θ
$\bar{O}$	132.27	34.90	2.73	53.21	44.76	3.99
$\bar{O}S_{1v}$+c.c.	20.15	5.25	−0.32	7.99	6.44	−0.44
$\bar{O}S_{2v}$+c.c.	31.21	7.90	−0.11	−78.62	12.89	−0.14
$S_{1v}^{\dagger}\bar{O}S_{1v}$	0.78	0.21	0.03	0.306	0.251	0.04
$S_{1v}^{\dagger}\bar{O}S_{2v}$+c.c.	2.08	0.22	−0.01	−2.81	0.34	−0.01
$S_{2v}^{\dagger}\bar{O}S_{2v}$+c.c.	8.81	−0.66	0.04	7.74	−1.13	0.05
Norm.	−4.03	−0.99	−0.05	0.24	−1.23	−0.07
Total	190.89	46.82	2.32	−11.99	62.27	3.42

Recently, calculated results of these properties have been reported by Itano using a multiconfiguration Dirac-Fock (MCDF) method [20]. The A and B results presented in that work are quite off from the experimental results. In fact, the result of A for the $5d\,^2D_{5/2}$ state has the opposite sign from the experimental result. We find that the difference between the Θ result of the $5d\,^2D_{3/2}$ state is due to different methods used, while the $5d\,^2D_{5/2}$ state result differs mainly due to the Breit interaction. In Table III, results obtained by both the methods are compared using the Dirac-Coulomb Hamiltonian. It demonstrates that the RCC theory is capable of considering correlation effects correctly in the present system and the Breit interaction contribution in the Θ result of

TABLE III. Different calculations of A, B, and Θ of the $5d$ states in Ba$^+$ using Dirac-Coulomb Hamiltonian.

	$5d\,^2D_{3/2}$			$5d\,^2D_{5/2}$		
	A	B	Θ	A	B	Θ
MCDF [20]	192.99	51.32	2.297	9.39	68.16	3.379
CCSD(T)	189.92	46.59	2.315	−11.72	62.69	3.382

the $5d\,^2D_{5/2}$ state is more than the $5d\,^2D_{3/2}$ state. These trends are different for the Θ calculation in ^{88}Sr$^+$ [38].

We also obtain the quadrupole polarizability $\alpha_2^0 = 1843.36 a_0^5$, with the $5d$ states using the following expression:

$$\alpha_2^0 = \frac{2}{5}\frac{1}{(2J_{6s}+1)^2}\sum_{n=5d_{3/2},5d_{5/2}} \frac{|\langle\Psi_{6s}\|\Theta_{zz}^{(2)}\|\Psi_n\rangle|^2}{E_n - E_{6s}}, \quad (14)$$

where the excitation energies are calculated using the CCSD(T) method (See Ref. [26]). This result is quite close to the experimental result $1828(88)a_0^5$ [39].

The most crucial part is to make a correct error analysis in the calculations of properties in many-electron systems. This is because, the calculations are carried out with a finite size of basis functions from a practical point of view and approximations in the method. We have carried out calculations with a different set of GTOs and considered the uncertainties from different results as one of the possible sources of errors. Other possible errors arise from the exclusion of higher order excitations by CCSD(T) method. It is very difficult to consider full triple and quadruple excitations in the heavy systems. It has been shown that methods such as unitary coupled-cluster method in the single and double approximations [UCCSD(T)] allow more leading order triple and quadruple excitations than the CCSD(T) method [40]. We consider all extra lowest order triple and quadruple excitation diagrams which arise from the UCCSD(T) method to estimate the error due to the CCSD(T) method. The difference of our calculations just with singles and doubles coupled-cluster (CCSD) method CCSD(T) method, which gives an impression of the role of the most leading order triple excitations, for Θ of the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states do not exceed contributions from the above discussed lowest order UCCSD(T) results. The extrapolated error bars from the above procedure to our calculated Θ values are around $0.02a_0^2$ and $0.04a_0^2$ for the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$, respectively.

In conclusion, we have employed the RCC theory to study both the magnetic dipole and electric quadrupole hyperfine structure constants rigorously in singly ionized barium. This study highlights the fact that accurate calculations for the d-state properties in heavy systems require a powerful method such as RCC theory to incorporate the electron core-polarization effects to all orders. We have also calculated quadrupole polarizability with $5d$ states and it is close to the recent experimental result. Using the wave functions that are able to produce the above compatible results with experiments, we obtain electric quadrupole moments for the $5d\,^2D_{3/2}$ and $5d\,^2D_{5/2}$ states as $2.32(2)ea_0^2$ and $3.42(4)ea_0^2$, respectively. These results will be used as testimony for the potential of the theory applied when experimental results will be known and will be used to extract out the Stark effects for finding the optical frequency standard with the single ionized barium.

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