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Polarizabilities and hyperfine structure constants of the low-lying levels of barium

M.G. Kozlov and S.G. Porsev[a]

Petersburg Nuclear Physics Institute, Gatchina, Leningrad district, 188350, Russia

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Abstract. The results of *ab initio* calculation of energies, hyperfine structure constants and static polarizabilities for several low-lying levels of barium are reported. The effective Hamiltonian for the valence electrons H_{eff} has been constructed in the frame of CI+MBPT method and solutions of many electron equation $H_{eff}\Phi_n = E_n\Phi_n$ were found. Using the wave functions obtained the hyperfine structure constants and static polarizabilities were calculated.

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1 Introduction

In this paper we report results of an *ab initio* calculation of static polarizabilities and hyperfine structure (hfs) constants for several low-lying levels of barium. In such calculations the accuracy of atomic wave function is tested twice, at large and short distances. Indeed, at short distances atomic wave function can be checked by the comparison of the calculated hfs constants with the experimental ones. The latter are usually known to a very good accuracy, providing a good test of the quality of the wave function near the nucleus.

Oscillator strengths and polarizabilities are, in contrast, determined by the wave function behavior at large distances. Usual experimental accuracy for the oscillator strengths and scalar polarizabilities is on the level of few percent. This is close or even less than the accuracy of precise atomic calculations (see, for example, calculations for Cs [1]). On the other hand, tensor polarizabilities can be measured with the accuracy of 1% or better (see, for instance, [2,3]). Thus, it is possible to test atomic wave function at large distances at 1% level. Note that 1% accuracy is crucial for calculations of parity non-conservation effects in atoms, because it allows to test predictions of the Standard model at small momentum transfer [1,4]. So far such precision has been achieved only for one-electron atoms Cs and Fr [5–7]. In this work we consider a much more complicated Ba atom.

We consider barium as a two electron atom with a xenon-like core. Valence-valence correlations are taken into account by configuration interaction (CI) method, while core-valence and core-core correlations are treated within the many body perturbation theory (MBPT). The latter is used to construct an effective Hamiltonian for the CI problem in the valence space. The details of the method can be found in the papers [8,9]. Application of this method to calculations of hfs constants has been discussed in [10]. Here we apply the same technique for calculations of atomic polarizabilities. In particular, we calculated scalar and tensor polarizabilities for five even-parity states $^1S_0(6s^2)$, $^3D_J(6s5d)$, and $^1D_2(6s5d)$ and four odd-parity states $^3P^o_J(6s6p)$ and $^1P^o_1(6s6p)$.

2 General formalism

Static polarizability of the sublevel $|a, J, M\rangle$ in a DC electric field $\mathbf{E} = \mathcal{E}\hat{\mathbf{z}}$ is defined as:

$$\begin{aligned}\Delta E_{a,J,M} &= -\frac{1}{2}\alpha_{a,J,M}\mathcal{E}^2 \\ &= -\frac{1}{2}\left(\alpha_{0,a,J} + \alpha_{2,a,J}\frac{3M^2 - J(J+1)}{J(2J-1)}\right)\mathcal{E}^2,\end{aligned} \tag{1}$$

where $\Delta E_{a,J,M}$ is an energy shift and α_0, α_2 define scalar and tensor polarizabilities correspondingly. Being a second order property, $\alpha_{a,J,M}$ can be expressed as a sum over unperturbed intermediate states:

$$\alpha_{a,J,M} = -2\sum_n \frac{|\langle a, J, M|D_z|n, J_n, M\rangle|^2}{E_a - E_n}, \tag{2}$$

where $\mathbf{D}$ is a dipole moment operator, E_n is an unperturbed energy of a level n, and the sum runs over all states of opposite parity. The formalism of the reduced matrix elements allows to write explicit expressions for the scalar

[a] e-mail: porsev@thd.pnpi.spb.ru

and tensor parts of the polarizability:

$$\alpha_{0,a,J} = \frac{-2}{3(2J+1)} \sum_n \frac{|\langle a,J||D||n,J_n\rangle|^2}{E_a - E_n}, \quad (3)$$

$$\alpha_{2,a,J} = 4\left(\frac{5J(2J-1)}{6(2J+3)(2J+1)(J+1)}\right)^{1/2} \times \sum_n (-1)^{J+J_n+1} \begin{Bmatrix} J & 1 & J_n \\ 1 & J & 2 \end{Bmatrix} \frac{|\langle a,J||D||n,J_n\rangle|^2}{E_a - E_n}, \quad (4)$$

and reduced matrix elements are defined as follows:

$$\langle n,J',M'|D_q|a,J,M\rangle = (-1)^{J'-M'} \begin{pmatrix} J' & 1 & J \\ -M' & q & M \end{pmatrix} \langle n,J'||D||a,J\rangle. \quad (5)$$

In order to use equations (2–4) in calculations one needs to know the complete set of eigenstates of the unperturbed Hamiltonian. It becomes practically impossible when dimension of a CI space exceeds few thousand determinants. It is known, that it is much more convenient to solve inhomogeneous equation instead of the direct summation over the intermediate states [11]. The general technique of the solution of an inhomogeneous equation was reported in [12].

Let us consider the solution of the following equation:

$$(E_a - H)|X_{a,M'}\rangle = D_q|a,J,M\rangle, \quad (6)$$

where $q = 0, \pm 1$ and $M' = M + q$. Obviously, the right hand side in (2) can be expressed in terms of the function $X_{a,M}$ (note that $r_0 \equiv r_z$):

$$\alpha_{a,J,M} = -2\langle a,J,M|D_0|X_{a,M}\rangle. \quad (7)$$

If we want to rewrite equations (3, 4) in terms of the function $X_{a,M'}$, we need to decompose the latter in terms, that correspond to particular angular momenta J_i. Generally speaking, there can be three such terms with $J_i = J, J \pm 1$:

$$X_{a,M'} = X_{a,J-1,M'} + X_{a,J,M'} + X_{a,J+1,M'}. \quad (8)$$

Now, with the help of the functions $X_{a,J',M'}$ the equations (3, 4) are reduced to:

$$\alpha_{0,a,J} = (-1)^{q+1} \frac{2}{3(2J+1)} \times \sum_{J'=J,J\pm 1} \begin{pmatrix} J' & 1 & J \\ -M' & q & M \end{pmatrix}^{-2} \langle a,J,M|D_{-q}|X_{a,J',M'}\rangle, \quad (9)$$

$$\alpha_{2,a,J} = 4(-1)^{q+1} \left(\frac{5J(2J-1)}{6(2J+3)(2J+1)(J+1)}\right)^{1/2} \times \sum_{J'=J,J\pm 1} (-1)^{J+J'} \begin{Bmatrix} J & 1 & J' \\ 1 & J & 2 \end{Bmatrix}^{-2} \begin{pmatrix} J' & 1 & J \\ -M' & q & M \end{pmatrix}^{-2} \times \langle a,J,M|D_{-q}|X_{a,J',M'}\rangle. \quad (10)$$

Note, that these equations are valid only if $3j$-symbols on the right hand side do not turn to zero. Thus, it is usually necessary to solve (6) for $q = \pm 1$, rather than for $q = 0$. Indeed, for $q = 0$ the $3j$-symbol that correspond, for instance, to $J' = J = 1$ and $M' = M = 0$, turns to zero.

If we know the solution of the equation (6) and its decomposition (8), then expressions (9, 10) allow us to find both scalar and tensor polarizabilities of the state $|a,J\rangle$. Moreover, the same functions $X_{a,J',M'}$ can be also used to find other second order atomic properties, such as amplitudes of Stark-induced $E1$ transitions or parity nonconserving $E1$ transitions between the states of the same nominal parity (see, for example [13]).

3 Calculation details

In this section we give a brief description of the calculation procedure putting special emphasis to the solution of the equation (6). Details of the construction of the effective Hamiltonian with the help of the MBPT diagrammatic technique can be found in [8,9]. Effective operators for hfs and dipole amplitudes include random phase approximation (RPA) corrections and several smaller MBPT corrections as described in [10].

3.1 Orbital basis set and CI space

This calculation is done in the V^N approximation, that means that core orbitals are obtained from the Dirac-Hartree-Fock (DHF) equations for a neutral atom (we use DHF computer code [14]). The basis set for the valence electrons includes $6s$, $6p$ and $5d$ DHF orbitals and $7s$–$16s$, $7p$–$16p$, $6d$–$13d$, $4f$–$12f$, and $5g$–$7g$ virtual orbitals. The latter were formed in two steps. On a first step we construct orbitals with the help of a recurrent procedure, which is similar to that suggested in [15] and described in [9,13]. After that we diagonalize the V^N DHF operator to obtain the final set of orbitals.

For the described orbital basis set the complete CI is made for both even-parity and odd-parity levels. Many electron wave functions are the linear combinations of the Slater determinants with a given J_z. That means that no symmetrization with respect to angular momentum J is made.

3.2 Effective operators

Within the CI+MBPT method the equation (6) is approximated by the equation in the valence space

$$(E_a - H_{eff})|X_{a,M'}\rangle = D_{eff,q}|a,J,M\rangle, \quad (11)$$

with the effective operators, which are found by means of the MBPT. The effective Hamiltonian for the two valence electrons is formed within the second order MBPT [8]. Only excitations of the 26 uppermost core electrons from

the shells $n = 4, 5$ were included. We used RPA for the effective dipole moment operator (see, for example, [16]). We have checked that MBPT corrections to $\mathbf{D}_{eff}$, which are not included in RPA are small, if RPA equations are solved in the V^{N-2} approximation, *i.e.* $6s$ electrons are excluded from the self-consistency procedure. The more detailed description of the effective operator formalism is given elsewhere [10].

In order to evaluate the right hand side of the equation (11), the matrix eigenvalue equation with the effective Hamiltonian has been solved for five low-lying even-parity states $^1S_0(6s^2)$, $^3D_J(6s5d)$, $J = 1, 2, 3$, and $^1D_2(6s5d)$ and four odd-parity states $^3P^o_J(6s6p)$, $J = 0, 1, 2$ and $^1P^o_1(6s6p)$.

3.3 Inhomogeneous equation

Technically there is no difference between equations (6, 11), and to simplify the notations we will speak about the solution of equation (6). This is a matrix equation with a large, sparse and symmetrical matrix. For the orbital basis set, in which DHF operator is diagonal, the nondiagonal matrix elements correspond to the residual two-electron interaction and are small. That allows to look for an iterative solution of the matrix equation (6). Let $X_a^{(n)}$ be an nth approximation and $R_a^{(n)}$ is the corresponding residue (here we omit quantum number M):

$$R_a^{(n)} = Y_a - (E_a - H)X_a^{(n)}, \tag{12}$$

where Y_a is the right hand side of (6). We now use the diagonal of the matrix H to form a probe vector $C_a^{(n)}$ and a new approximate solution $X_a^{(n+1)}$:

$$C_a^{(n)} = (E_a - \mathrm{diag}(H))^{-1} R_a^{(n)}, \tag{13}$$

$$X_a^{(n+1)} = k_1 X_a^{(n)} + k_2 C_a^{(n)} + \sum_{l=1}^{N} k_{l+2}\Psi_l, \tag{14}$$

where coefficients k_i are found from a minimum residue condition and Ψ_l are some (approximate) eigenfunctions of the effective Hamiltonian.

The procedure (12–14) provides the monotonous decrease of the residue. For the ground state ($a = 0$), this procedure converges rather rapidly even for $N = 0$. For excited states convergence can be significantly slowed down when there are eigenvalues of the effective Hamiltonian, which are close to E_a. Suppose that there are N eigenfunctions of the opposite parity Ψ_l and $|E_l - E_a| \ll 1$, $l = 1, \ldots N$. We can speed up the convergence by adding these functions to the right hand side of (14) and find a minimum of the residue with respect to all $N + 2$ coefficients k_i. Note that it does not require any additional time-consuming computations because the product $H\Psi_l$ has to be calculated only once. Such iterative procedure works fine, provided that all most important eigenvectors and eigenvalues have been found beforehand.

Table 1. Energy levels for Ba in V^N approximation.

Level	E_{val} (a.u.)	Δ (cm^{-1})	Experiment (cm^{-1})[a]
$^1S_0$	−0.55916[b]	0	0
$^3D_1$	−0.51786	9064	9034
$^3D_2$	−0.51700	9254	9216
$^3D_3$	−0.51518	9653	9597
$^1D_2$	−0.50678	11496	11395
$^3P^o_0$	−0.50319	12284	12266
$^3P^o_1$	−0.50150	12656	12637
$^3P^o_2$	−0.49750	13533	13515
$^1P^o_1$	−0.47600	18252	18060

[a] Reference [21].
[b] The absolute value of this energy corresponds to the sum of the two first ionization potentials of barium, for which experiment gives 0.559148 a.u. [21].

3.4 Decomposition of the function $X_{a,M'}$

When the function $X_{a,M'}$ is found, the decomposition (8) is easily done with the help of the following operators $\hat{P}_i$:

$$X_{a,J_i,M'} = \hat{P}_i X_{a,M'}, \tag{15}$$

$$\hat{P}_i = N_i \left(\hat{\mathbf{J}}^2 - J_k(J_k+1)\right)\left(\hat{\mathbf{J}}^2 - J_l(J_l+1)\right), \tag{16}$$

$$N_i = (J_i(J_i+1) - J_k(J_k+1))^{-1} \times (J_i(J_i+1) - J_l(J_l+1))^{-1},$$

where $\hat{\mathbf{J}}$ is the angular momentum operator and $J_i \neq J_k \neq J_l \in \{J-1, J, J+1\}$.

Let us point out that the operator $\hat{P}_i$ is not a projector on a subspace with $J = J_i$, but it works as such when applied to a three component function like $X_{a,M'}$. Indeed, $\hat{P}_i$ eliminates components with $J_k \neq J_i$, $J_k \in \{J-1, J, J+1\}$ and leaves the component with $J_k = J_i$ unchanged. If $|a, J, M\rangle$ in the right hand side in (6) is not an exact eigenvector of the operator H, or $X_{a,M'}$ is not an exact solution of the equation (6), then the latter can have admixtures with other angular momenta. In this case functions defined by (15) are not eigenfunctions of $\hat{\mathbf{J}}^2$ and are not orthogonal to each other. This provides us with a simple check whether the function $X_{a,M'}$ can be decomposed in three components with definite angular momenta or not. It is sufficient to check normalization of the right hand side and the left hand side of the equation (8). So, operators $\hat{P}_i$ allow not only to decompose the function $X_{a,M'}$ in terms with definite angular momenta, but also to check the quality of this function.

4 Results and discussion

Results of the solution of the eigenvalue problems for even-parity and odd-parity levels are presented in Table 1.

Table 2. Magnetic dipole (A) and electric quadrupole (B) hyperfine structure constants of low-lying levels for ^{137}Ba. The electric quadrupole moment is taken to be 0.245 b.

	$^3D_1$	$^3D_2$	$^3D_3$	$^1D_2$	$^3P_1^o$	$^3P_2^o$	$^1P_1^o$
				A (MHz)			
Dirac-Fock	−430	267	402	38	724	594	75
CI	−353	306	350	− 25	794	595	− 46
CI + MBPT	−547	405	443	−102	1160	845	−107
Experiment	−521[a]	416[a]	457[a]	− 82[b]	1151[c]	—	−109[d]
Theory (MCDF) [20]	—	320	—	− 13	804	580	− 53
				B (MHz)			
Dirac-Fock	12.7	17.7	32.2	33.5	−23.2	40.0	43.2
CI	12.5	18.1	31.9	39.7	−26.6	47.0	26.8
CI + MBPT	17.7	26.8	47.4	67.2	−43.2	77.4	58.4
Experiment	17.9[a]	25.9[a]	47.4[a]	59.6[b]	−41.6[c]	—	51[d]
Theory (MCDF) [20]	—	47.9	—	40.8	52.5	61.4	−7.7

[a][22]; [b][23]; [c][24]; [d][25].

These results are close to our earlier calculations [9] and to the results of the recent paper [17], where the same method was used. The typical accuracy for the energy intervals is better than 100 cm^{-1} with the only exception of $^1P_1^o$ level, where the error is almost 200 cm^{-1}. Note, that typical error of the coupled-cluster calculation of the same levels of Ba is $200 \div 400$ cm^{-1} [18]. In [17] it was shown, that agreement with the experiment can be further improved if higher order MBPT corrections to the effective Hamiltonian are included semiempirically. In particular, in the second order of MBPT there is a large screening of exchange interaction between $6s$ and $6p$ electrons, which affects the splitting between states $^3P_J^o$ and $^1P_1^o$. Higher order corrections to this screening should be essential [19].

When the eigenvalue problem is solved, we can calculate the hfs constants A and B (Tab. 2) and the polarizabilities of the corresponding states (Tab. 3). On the CI stage our results for the constant A are close to those of the paper [20], where the multi-configurational Dirac-Fock (MCDF) method was used. It is seen that these results strongly underestimate magnetic hfs for all levels. On average, MBPT corrections contribute $30\% \div 50\%$ and even more for small constants. The final accuracy of the theory for the constant A appears to be about $3\% \div 5\%$ (except $^1D_2$-state), which is somewhat less than the accuracy of the similar calculations for Tl [10]. It is probably the consequence of the small numerical values of hfs constants for Ba. Indeed, the largest hfs constant of the level $^3P_1^o$ is calculated with the highest accuracy. For other levels there are cancelations between contributions from $6s$-electron and either $5d$- or $6p$-electron. We have not found experimental data for the level $^3P_2^o$. MBPT corrections for this level are of the typical size, cancelations of different contributions are moderate, so we estimate the accuracy of the theory for this level to be about 3% or better.

Similar situation takes place for the hfs constant B, but here our results are significantly different from those of [20] already on the CI stage. The role of the MBPT corrections is even higher here. These corrections almost double the answer for the levels $^1D_2$ and $^1P_1^o$. So, it is not surprise, that the accuracy of the theory for these levels is not very high. Again, for the level $^3P_2^o$ the constant B is unknown. In this case MBPT corrections contribute more than 60%, so we can not guarantee the accuracy higher than 15%.

Table 3. Scalar and tensor polarizabilities (a.u.) of low-lying levels for Ba.

		Experiment		Theory	
conf.	level	α_0	α_2	α_0	α_2
$6s^2$	^{1}S$_0$	268(22)[a]	0	264	0
$6s5d$	$^3D_1$	$X + 16(4)$[b]	−53(1)[b]	383	− 60
$6s5d$	$^3D_2$	$X + 4(4)$[b]	−69(1)[b]	372	− 79
$6s5d$	$^3D_3$	X	−121(4)[b]	365	−141
$6s5d$	$^1D_2$	$X - 109(44)$[b]	86(2)[b]	266	81
$6s6p$	$^3P_0^o$		0	− 13	0
$6s6p$	$^3P_1^o$		0.169(28)[c]	− 10	0.9
$6s6p$	$^3P_2^o$			62	− 11
$6s6p$	$^1P_1^o$	411(23)[d]	− 43.1(4)[e]	409	− 51

[a] Reference [26];
[b] reference [27], scalar polarizabilities in this paper were measured relative to that of the level $^3D_3$, which is denoted by X;
[c] reference [28];
[d] reference [29];
[e] reference [2].

In order to find polarizabilities we substitute eigenfunctions in equation (11) and solve corresponding inhomogeneous equation. After that equations (9, 10) give us α_0 and α_2. Results of these calculations are listed

in Table 3. Scalar polarizabilities for $^1S_0$- and $^3P_0^o$-levels are known from the experiment. For the D-states scalar polarizabilities were measured relative to that of the level $^3D_3$. In Table 3 the latter is designated by X. If we assume that X is equal to the corresponding theoretical value, all experimental data for α_0 agree with our calculations within the experimental uncertainty.

It is seen that typically α_2 is about an order of magnitude smaller than α_0. This is due to the strong cancellations between three terms of the sum (10). For this reason the theoretical accuracy for α_2 is significantly lower than for α_0. On the contrary, experimental data for α_2 are much more precise.

There are two main sources of errors in the calculations of polarizabilities. First one is the same as for the hfs calculations, and connected with the inaccuracy in the wave functions and effective operators (note that RPA corrections to the dipole operator are much smaller than for hfs operators). Second source of errors is the inaccuracy in eigenvalues, that is specific to the calculations of polarizabilities. Normally, the errors associated with the latter are small, but they can become important (and even dominant) when there are close levels of opposite parity. In particular, that applies to levels $^1D_2$ and $^3P_1$, and, to some extent, to other levels of configurations $6s5d$ and $6s6p$. Indeed, the energy interval between levels $^1D_2$ and $^3P_1$ is underestimated by 7% (see Tab. 1). This interval enters equations (1, 2) and potentially can cause a large error. Fortunately, corresponding numerators are small, because singlet-triplet amplitudes are suppressed. On the other hand, this error is relatively enhanced for the tensor polarizability, because of the strong cancellations of different contributions, mentioned above.

In conclusion let us sum up the results. We have found approximate solutions of many electron equation $H_{eff}\Phi_n = E_n\Phi_n$ for low-lying levels of barium. Then, we tested the quality of the wave functions obtained at large and short distances. To do this, we calculated hfs constants and polarizabilitilies for the levels in question. Calculations of polarizabilities of the excited states were hampered due to the proximity of the levels of opposite parity. Further improvement of theoretical accuracy for these polarizabilities requires better agreement between theoretical and experimental spectrum of the atom. That can be done, if higher-order MBPT corrections to the effective Hamiltonian are included semiempirically [17,19].

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