

## Electric-dipole amplitudes, lifetimes, and polarizabilities of the low-lying levels of atomic ytterbium

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The results of *ab initio* calculations of electric-dipole amplitudes, lifetimes, and polarizabilities for several low-lying levels of ytterbium are reported. The effective Hamiltonian for two valence electrons  $H_{\text{eff}}$  was constructed by means of the many-body perturbation theory and solutions of the two-electron equation  $H_{\text{eff}}\Phi_n = E_n\Phi_n$  were found. [S1050-2947(99)09307-5]

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### I. INTRODUCTION

In this paper we report results of an *ab initio* calculation of electric-dipole ( $E1$ ) amplitudes, lifetimes, and polarizabilities for several low-lying levels of ytterbium. In Ref. [1] we calculated the energies and hyperfine structure (hfs) constants of low-lying levels of ytterbium. In that calculation the accuracy of the atomic wave functions was tested by comparison of the calculated hfs constants with the experimentally determined 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.

$E1$  amplitudes, in contrast, are determined by the form of the wave function at large distances. The usual experimental accuracy for the oscillator strengths and scalar polarizabilities is on the level of a few percent. This is close to, or even less than, the accuracy of precise atomic calculations (see, e.g., calculations for Ba [2] and Cs [3]). Tensor polarizabilities can be measured to an accuracy of 1% or better [4,5]. Thus it is possible to test an atomic wave function at large distances at the 1% level. Note that 1% accuracy is crucial for calculations of parity nonconservation effects in atoms because it allows predictions of the Standard model to be tested in the regime of small momentum transfer [3,6]. To date, such precision has been achieved only for the one-electron atoms Cs and Fr [7–9]. In this paper we deal with the much more complicated Yb atom.

We consider ytterbium as a two-electron atom with the core  $[1s^2, \dots, 4f^{14}]$ . Valence-valence correlations are taken into account by the configuration interaction (CI) method, while core-valence and core-core correlations are treated within the second order of the many-body perturbation theory (MBPT). The latter is used to construct an effective Hamiltonian for the CI problem in the valence space. (Details of the method can be found elsewhere [10,11]). Application of this method to the calculation of hfs constants has been discussed in [1,2,12]. In Ref. [2] the method was extended to the calculation of polarizabilities. Here we apply this technique for calculating lifetimes,  $E1$  amplitudes, and polarizabilities of ytterbium.

### II. GENERAL FORMALISM

The expression for the oscillator strength for an  $a, J \rightarrow a', J'$  transition has the form [13] (atomic units  $m = \hbar$

$= e = 1$  are used throughout the paper):

$$f(aJ, a'J') = -\frac{2\omega_{aJ, a'J'}}{3(2J+1)} |\langle a, J || D || a', J' \rangle|^2, \quad (1)$$

where  $\omega_{aJ, a'J'} = E_{aJ} - E_{a'J'}$ ,  $\mathbf{D}$  is the dipole moment operator, and reduced matrix elements (MEs) are defined as follows:

$$\begin{aligned} \langle a', J', M' | D_q | a, J, M \rangle &= (-1)^{J'-M'} \begin{pmatrix} J' & 1 & J \\ -M' & q & M \end{pmatrix} \\ &\times \langle a', J' || D || a, J \rangle. \end{aligned} \quad (2)$$

The lifetime  $\tau$  of a level is the inverse of the total transition rate. The probability for an  $a, J \rightarrow a', J'$  transition is given by

$$W(aJ, a'J') = \frac{4}{3c^3} \frac{\omega_{aJ, a'J'}^3}{2J+1} |\langle a, J || D || a', J' \rangle|^2, \quad (3)$$

where  $c$  is the speed of light.

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

$$\begin{aligned} \Delta E_{a, J, M} &= -\frac{1}{2} \alpha_{a, J, M} \mathcal{E}^2 \\ &= -\frac{1}{2} \left( \alpha_{0, a, J} + \alpha_{2, a, J} \frac{3M^2 - J(J+1)}{J(2J-1)} \right) \mathcal{E}^2, \end{aligned} \quad (4)$$

where  $\Delta E_{a, J, M}$  is the energy shift and  $\alpha_0$  and  $\alpha_2$  define the scalar and tensor polarizabilities, respectively. The polarizability  $\alpha_{a, J, M}$  can be expressed as a sum over unperturbed intermediate states:

$$\alpha_{a, J, M} = -2 \sum_n \frac{|\langle a, J, M | D_z | n, J_n, M \rangle|^2}{E_a - E_n}, \quad (5)$$

where  $E_n$  is an unperturbed energy of a level  $n$ , and the sum runs over all states of opposite parity. The formalism of the reduced MEs allows us to write explicit expressions for the scalar and tensor parts of the polarizability:

$$\alpha_{0,a,J} = \frac{-2}{3(2J+1)} \sum_n \frac{|\langle a,J||D||n,J_n\rangle|^2}{E_a - E_n}, \quad (6)$$

$$\alpha_{2,a,J} = \left( \frac{40J(2J-1)}{3(2J+3)(2J+1)(J+1)} \right)^{1/2} \sum_n (-1)^{J+J_n+1} \times \begin{Bmatrix} J & 1 & J_n \\ 1 & J & 2 \end{Bmatrix} \frac{|\langle a,J||D||n,J_n\rangle|^2}{E_a - E_n}. \quad (7)$$

In order to use Eqs. (5)–(7) in calculations one needs to know a complete set of eigenstates of the unperturbed Hamiltonian. It becomes practically impossible when the dimension of a CI space exceeds a few thousand. For such cases, however, it is known that it is much more convenient to solve the inhomogeneous equation instead of the direct summation over the intermediate states [14,15]. Therefore, let us consider the solution of the following inhomogeneous equation:

$$(E_a - H)|X_{a,M'}\rangle = D_q|a,J,M\rangle, \quad (8)$$

where  $q=0,\pm 1$  and  $M'=M+q$ . Obviously, the right-hand side in Eq. (5) can be expressed in terms of the function  $X_{a,M}$  (note that  $D_0 \equiv D_z$ ):

$$\alpha_{a,J,M} = -2\langle a,J,M|D_0|X_{a,M}\rangle. \quad (9)$$

If we want to rewrite Eqs. (6) and (7) 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  $J_i = J, J \pm 1$ :

$$X_{a,M'} = X_{a,J-1,M'} + X_{a,J,M'} + X_{a,J+1,M'}. \quad (10)$$

Now, with the help of the functions  $X_{a,J',M'}$ , Eqs. (6) and (7) reduce to

$$\alpha_{0,a,J} = (-1)^{q+1} \frac{2}{3(2J+1)} \times \sum_{J'} \begin{pmatrix} J' & 1 & J \\ -M' & q & M \end{pmatrix}^{-2} \langle a,J,M|D_{-q}|X_{a,J',M'}\rangle, \quad (11)$$

$$\alpha_{2,a,J} = (-1)^{q+1} \left( \frac{40J(2J-1)}{3(2J+3)(2J+1)(J+1)} \right)^{1/2} \times \sum_{J'} (-1)^{J+J'} \begin{Bmatrix} J & 1 & J' \\ 1 & J & 2 \end{Bmatrix}^{-2} \begin{pmatrix} J' & 1 & J \\ -M' & q & M \end{pmatrix}^{-2} \times \langle a,J,M|D_{-q}|X_{a,J',M'}\rangle, \quad (12)$$

with summations running over  $J' = J, J \pm 1$ . Note that these equations are valid only if all  $3j$  symbols on the right-hand side are not zero. This constraint must be taken into account when choosing for what spherical component  $q$  to solve Eq. (8).

If we know the solution of Eq. (8) and its decomposition (10), then expressions (11) and (12) allow us to find both the scalar and the tensor polarizabilities of the state  $|a,J\rangle$ . Moreover, the same functions  $X_{a,J',M'}$  can also be used to find

other second-order atomic properties, such as amplitudes of the Stark-induced  $E1$  transitions, or parity nonconserving  $E1$  transitions, between the states of the same nominal parity (see, for example, Ref. [16]).

### III. CALCULATION DETAILS AND RESULTS

#### A. Orbital basis set and CI space

The calculation procedure is quite similar to that of Ref. [2]; therefore, we give here only a brief description of its features. The calculation is done in the  $V^N$  approximation, i.e. the core orbitals are obtained from the Dirac-Hartree-Fock (DHF) equations for a neutral atom (we use the DHF computer code [17]). The basis set for the valence electrons includes  $6s, 6p, 5d, 7s, 7p$ , and  $6d$  DHF orbitals and  $8s-15s, 8p-15p, 7d-14d, 5f-10f$ , and  $5g-7g$  virtual orbitals. The latter were formed in two steps. On the first step we construct orbitals with the help of a recurrent procedure, which is similar to that suggested in Ref. [18] and described in Refs. [11,16]. Subsequently we diagonalize the  $V^N$  DHF operator to obtain the final set of orbitals.

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

#### B. Effective operators

Within the CI+MBPT method the wave function of the valence electrons is found from the eigenvalue equation:

$$H_{\text{eff}}|a,J,M\rangle = E_a|a,J,M\rangle. \quad (13)$$

Equation (8) is rewritten as an equation for valence electrons only:

$$(E_a - H_{\text{eff}})|X_{a,M'}\rangle = D_{\text{eff},q}|a,J,M\rangle, \quad (14)$$

with the effective operators, which are found by means of the MBPT. The effective Hamiltonian for two valence electrons is formed within the second-order MBPT [10]. We used the random-phase approximation (RPA) for the effective dipole moment operator (see, for example, Ref. [19]). We have checked that MBPT corrections to  $\mathbf{D}_{\text{eff}}$ , which are not included in the RPA, are small if the RPA equations are solved with the  $6s$  electrons excluded from the self-consistency procedure (i.e., the RPA equations have the same form as in the  $V^{N-2}$  approximation). A more detailed description of the effective operator formalism is given in [12].

#### C. Transition amplitudes and lifetimes

We first solve eigenvalue Eq. (13) with the effective Hamiltonian for low-lying even- and odd-parity states. Strictly speaking, the effective Hamiltonian can be safely used only for the energy levels below the core excitation threshold. For Yb this threshold lies at  $23189 \text{ cm}^{-1}$  above the ground state [20]. However, it was shown in [1] that the theoretical spectrum is quite good up to  $\sim 40000 \text{ cm}^{-1}$ . Consequently, we can work (with some caution) with the states lying slightly above the core-excitation threshold. Our approach fails to reproduce the states with an unfilled  $f$  shell

TABLE I. Reduced MEs  $|\langle L_J || r || L'_J \rangle|$  (a.u.). Calculations were made in the  $L$  gauge. Other theoretical and experimental results are given where available. The uncertainties are indicated in parentheses. Dashes indicate that the corresponding transitions are forbidden.

	${}^3P_0^o(6s6p)$	${}^3P_1^o(6s6p)$	${}^3P_2^o(6s6p)$	${}^1P_1^o(6s6p)$
${}^1S_0(6s^2)$	—	0.54 (8) 0.44 <sup>a</sup> 0.549 (4) <sup>c</sup> 0.553 (13) <sup>d</sup>	—	4.40 (80) 4.44 <sup>a</sup> 4.89 <sup>b</sup> 4.13 (10) <sup>d</sup> 4.02 <sup>e</sup> 4.26 <sup>f</sup>
${}^3D_1(5d6s)$	2.61 (10)	2.26 (10) 2.2 (1) <sup>g</sup>	0.60 (12)	0.27 (10) 0.24 <sup>a</sup>
${}^3D_2(5d6s)$	—	4.03(16)	2.39 (10)	0.32 (6) 0.60 <sup>a</sup>
${}^3D_3(5d6s)$	—	—	6.12 (30)	—
${}^1D_2(5d6s)$	—	0.54 (10)	0.38 (8)	3.60 (70)
${}^3S_1(6s7s)$	1.98 (10) 1.36 <sup>b</sup>	3.53 (15) 2.50 <sup>b</sup>	5.05 (20) 3.77 <sup>b</sup>	0.73 (15)
${}^1S_0(6s7s)$	—	0.22 (4) 0.22 (2) <sup>h</sup>	—	4.31 (80)

<sup>a</sup>Reference [22].

<sup>b</sup>Reference [21] (footnotes a and b are theory).

<sup>c</sup>Reference [23].

<sup>d</sup>Reference [24].

<sup>e</sup>Reference [25].

<sup>f</sup>Reference [26].

<sup>g</sup>Reference [27].

<sup>h</sup>Reference [28] (footnotes c–h are from experiment).

or to account properly for the interaction with such states. For this reason we consider only the states, which lie sufficiently far from those with an unfilled  $f$  shell. In particular, we consider  $E1$  transitions between four low-lying odd-parity states [ ${}^3P_{0,1,2}^o(6s6p)$  and  ${}^1P_1^o(6s6p)$ ] and seven even-parity states [ ${}^1S_0(6s^2)$ ,  ${}^3D_{1,2,3}(5d6s)$ ,  ${}^1D_2(5d6s)$ ,  ${}^3S_1(6s7s)$ , and  ${}^1S_0(6s7s)$ ]. The state  ${}^1P_1^o(6s6p)$  requires special attention. The nearest  $f^{13}5d6s^2$  state lies only  $3800 \text{ cm}^{-1}$  above the latter and their interaction is not negligible. We estimated that the configuration  $f^{13}5d6s^2$  contributes on the level of several percent to the wave function of  ${}^1P_1^o(6s6p)$  state. The accuracy of the calculated  ${}^1P_1^o(6s6p) \rightarrow {}^1L_J$  and  $E1$  amplitudes is reduced because we do not take into account this configuration mixture.

After the eigenfunctions for the valence electrons are found, we can calculate transition amplitudes and lifetimes. The results of these calculations are presented in Table I. The magnitudes of the  $E1$  amplitudes vary over a wide range. These variations correspond in part to the approximate selection rules  $\Delta S=0$  and  $\Delta J=\Delta L$ , which are easily traced through Table I. For large amplitudes we estimate the accuracy of our calculation to be 3–5%. For the reason discussed above the amplitudes  $\langle {}^1L_J || D || {}^1P_1^o(6s6p) \rangle$  do not follow this rule. The accuracy for these amplitudes, as well as for small amplitudes ( $\leq 0.5$  a.u.), is about 15–20%.

A few of these reduced MEs have been calculated previously [21,22] or determined experimentally [23–28], and we

TABLE II. Lifetimes (nsec) of the low-lying levels for Yb calculated with the reduced MEs from Table I and experimental transition frequencies.

State	Config.	This work	Other data
${}^3D_1$	$5d6s$	372 (30)	380 (30) <sup>a</sup>
${}^3D_2$	$5d6s$	430 (35)	460 (30) <sup>a</sup>
${}^3D_3$	$5d6s$	540 (55)	
${}^1D_2$	$5d6s$	4400 (1800)	6700 (500) <sup>a</sup>
${}^3S_1$	$6s7s$	13.5 (1.1)	12.5 (1.5) <sup>b</sup> 15.9 (1.9) <sup>c</sup>
${}^1S_0$	$6s7s$	33 (13)	45.8 (1.0) <sup>d</sup>
${}^3P_1^o$	$6s6p$	875 (250)	760–875 <sup>e</sup> 1294 <sup>f</sup>
${}^3P_2^o$	$6s6p$	15.0 (1.5) sec	14.5 sec <sup>f</sup>
${}^1P_1^o$	$6s6p$	5 (2)	5.1–6.4 <sup>e</sup> 4.78 <sup>f</sup>

<sup>a</sup>Reference [27].

<sup>b</sup>Reference [33].

<sup>c</sup>Reference [34].

<sup>d</sup>Reference [28].

<sup>e</sup>See Ref. [35] and references therein (footnotes a–e are from experiment).

<sup>f</sup>Reference [22] (theory).

compare these with our results in Table I. For the convenience of comparison we used Eqs. (1) and (3) to calculate reduced MEs from the oscillator strengths and transition probabilities. The calculations in Ref. [21] were performed in the  $L$ - $S$  coupling scheme and the simplest semiempirical method [29] was then used to evaluate the radial parts. In Ref. [22] the multiconfiguration Dirac-Fock method was used, with the valence-core electronic correlations included semiempirically. Comparing our results with the results of other theoretical works highlights the importance of accounting for the valence-core correlations.

Now, using Eq. (3), we can find the transition probabilities and the lifetimes of the levels (see Table II). In these calculations we used experimental transition frequencies. Therefore, the accuracy of these numbers depends only on the accuracy of the dominant transition amplitudes. As a result, the largest error (40%) takes place for the states  ${}^1S_0(6s7s)$  and  ${}^1D_2(5d6s)$  where the transition to the state  ${}^1P_1^o(6s6p)$  is dominant. For other states we estimate the theoretical accuracy for the lifetimes to be 10% or better.

#### D. Polarizabilities

In order to find the polarizabilities we substitute eigenfunctions into the right-hand side of Eq. (14) and solve the corresponding inhomogeneous equation. Then, use of Eqs. (11) and (12) enables us to calculate  $\alpha_0$  and  $\alpha_2$ . The results of these calculations are presented in Table III. It is seen that  $\alpha_2$  has typically the same order of magnitude as  $\alpha_0$  (in contrast to the case of barium [2]). Therefore, the theoretical accuracy for  $\alpha_2$ , as a rule, is similar to that for  $\alpha_0$ . In contrast, experimental data for  $\alpha_2$  are usually much more precise and complete.

There are several sources of errors in the calculations of polarizabilities. Some of them are the same as for hfs calcu-

TABLE III. Scalar and tensor polarizabilities (a.u.) of low-lying levels of Yb. Theoretical accuracy is indicated where analysis was possible; otherwise, the numbers should be considered as estimates.

Level	Config.	Theory		Experiment
		$\alpha_0$	$\alpha_2$	$\alpha_2$
$^1S_0$	$6s^2$	118 (45)		
$^3D_1$	$5d6s$	47	22	28 (4) <sup>a</sup>
$^3D_2$	$5d6s$	36	17	28 (8) <sup>a</sup>
$^3D_3$	$5d6s$	-9	118 (24)	
$^1D_2$	$5d6s$	4	150	
$^1S_0$	$6s7s$	2072		
$^3S_1$	$6s7s$	2030	0.8	
$^3P_0^o$	$6s6p$	252 (25)		
$^3P_1^o$	$6s6p$	278 (15)	24.3 (1.5)	24.06 (1.37) <sup>b</sup> 24.26 (0.84) <sup>c</sup> 23.33 (0.52) <sup>d</sup>
$^3P_2^o$	$6s6p$	383 (32)	-76(6)	
$^1P_1^o$	$6s6p$	501 (200)	-118(60)	-57.4(5.6) <sup>b</sup>

<sup>a</sup>Reference [23].

<sup>b</sup>Reference [36].

<sup>c</sup>Reference [37].

<sup>d</sup>Reference [31].

lations, and are connected with the inaccuracy in the wave functions and the effective operators (note that the RPA corrections to the dipole operator are much smaller than for hfs operators). The additional source of error is the inaccuracy in eigenvalues. Finally, solving Eq. (14) we did not account for the configurations  $4f^{13}nl'n''l''$ . Fortunately, the states from these configurations that can be reached by one-electron transitions from the levels studied here have large energies. The estimates show that the contribution of  $f$ -shell polarization to the polarizabilities of the states listed in Table III does not exceed 2–3 a.u.

The final accuracy of the calculations is very different for different levels. For instance, 95% of the polarizability of the ground state  $^1S_0(6s^2)$  is due to the ME  $\langle ^1S_0 | D | ^1P_1^o(6s6p) \rangle$ . Supposing that this ME is calculated with an accuracy of 20%, the accuracy for  $\alpha_0(^1S_0)$  will be about 40% (the corresponding transition frequency is reproduced almost ideally [1]). It should be pointed out that even taking into account the large uncertainty of our result, it significantly differs from  $\alpha_0(^1S_0)=266$  a.u. obtained in Ref. [30] where the Hartree-Fock method was used.

For the  $D_J(5d6s)$  states the situation is more complicated. There are large cancellations between the contributions of  $P_J^o(6s6p)$  states and higher-lying states. For this reason their polarizabilities are small and the role of different small contributions is enhanced. Thus, analysis of the accuracy becomes difficult; only for the tensor polarizability of  $^3D_3(5d6s)$  state we can estimate the accuracy to be 20%. All other values of  $\alpha_0$  and  $\alpha_2$  for the  $D_J(5d6s)$  states presented in Table III are order of magnitude estimates.

The scalar polarizabilities of the levels  $^3S_1(6s7s)$  and  $^1S_0(6s7s)$  are basically determined by the MEs  $\langle S_{J'}(6s7s) | D | P_J^o(6s7p) \rangle$ . Because of the closeness of the  $f^{13}5d^26s$  states we failed to obtain reliable wave functions for the  $P_J^o(6s7p)$  states. Consequently, the values for

$\alpha_0(^3S_1(6s7s))$  and  $\alpha_0(^1S_0(6s7s))$  are also only order of magnitude estimates.

Now let us consider the odd-parity states. The accuracy of  $\alpha_0$  and  $\alpha_2$  for the  $^3P_J^o(6s6p)$  triplet is 6–10%. The main contribution here comes from the  $^3D_J(5d6s)$  multiplet and there are no cancellations because all of the important levels of opposite parity lie above and hence contribute with the same sign. The accuracy for  $\alpha_0$  of the  $^1P_1^o(6s6p)$  state is about 40% and is even worse (50%) for  $\alpha_2$ . This is a consequence of the large contribution to these polarizabilities from the intermediate state  $^1S_0(6s7s)$  (see above).

In Ref. [31] the Stark shift of the  $^1S_0(6s^2) \rightarrow ^3P_1^o(6s6p)$  transition in ytterbium was measured. The Stark shift rate was found to be  $K = -61.924(0.193)$  a.u. In terms of polarizabilities it can be written as:

$$K = -\frac{1}{2}\{\alpha_0(^3P_1^o) - 2\alpha_2(^3P_1^o) - \alpha_0(^1S_0)\}.$$

Using the numbers from Table III, we find that  $K = -55(9)$  a.u., in good agreement with the experimental result [31].

In Ref. [23], the Stark shifts for the  $^1S_0(6s^2) \rightarrow ^3D_{1,2}(5d6s)$  transitions were observed. These shifts depend on the differences in scalar polarizabilities ( $\alpha_0(^1S_0) - \alpha_0(^3D_{1,2})$ ):

$$\alpha_0(^1S_0) - \alpha_0(^3D_1) = \begin{cases} 71 & \text{theory} \\ 86 (3) & \text{experiment,} \end{cases}$$

$$\alpha_0(^1S_0) - \alpha_0(^3D_2) = \begin{cases} 82 & \text{theory} \\ 80 (4) & \text{experiment,} \end{cases}$$

where theoretical values are taken from Table III.

The method used here allows us to calculate not only static polarizabilities, but also the Stark-induced amplitudes

for different transitions. For instance, the magnitude of the vector-transition polarizability  $|\beta|$  for the  $^1S_0(6s^2) \rightarrow ^3D_1(5d6s)$  transition was calculated to be 122 (12) a.u., in good agreement with our previous calculation 138 (30) a.u. [32] and experimental result 113 (15) a.u. [23].

#### IV. CONCLUSION

Application of the effective operator technique to the Yb atom is hampered by the existence of the shallow  $4f$  shell. Nevertheless, it is possible to make reliable calculations of different atomic properties including transition frequencies, hyperfine constants,  $E1$  amplitudes, lifetimes, and polariz-

abilities for many low-lying energy levels. It is of a particular importance that, with some caution, calculations can be done even for levels that lie above the core-excitation threshold, which is at  $23189 \text{ cm}^{-1}$ .

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