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Lifetime measurements and calculations in singly ionized ytterbium

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Abstract

New radiative lifetimes, measured by time-resolved laser-induced fluorescence spectroscopy, are reported for five Rydberg states of singly ionized ytterbium. Free Yb⁺ ions were produced in a laser-induced plasma. The experimental results have been compared with HFR calculations, taking core-polarization effects into account, and a good agreement (within 25%) between theory and experiment is observed for four levels. HFR results are also proposed for np ($n \leq 7$) and nf ($n \leq 8$) Rydberg states and compared with available data.

1. Introduction

The ytterbium ion has attracted much attention from physicists in recent years for several reasons. It has special interest for atomic clocks and trapped-ion frequency standards in relation to the structure of the low-lying levels giving rise to optical, infrared or microwave frequency standards (Tamm *et al* 1995, Fisk *et al* 1995, Gill *et al* 1995, Roberts *et al* 1997, Taylor *et al* 1998). Among the transitions of particular interest, the electric octupole $^2S_{1/2} - ^2F_{7/2}$ 467 nm transition, belonging to the isotopes 172 or 171 of Yb⁺, which has a lifetime of around 10 years, has attracted special attention, both experimentally (Roberts *et al* 1997, 2000) and theoretically (Biémont and Quinet 1998). The lifetimes of the $4f^{14}5d^2D_{3/2}$ and $^2D_{5/2}$ levels, usable as a basis for an optical frequency standard, have recently been investigated by Yu and Maleki (2000). The Yb⁺ ion is also well suited for cavity QED studies and for quantum computation tests with the help of trapped ions (Yu and Maleki 2000).

Up to now, the lifetime values measured in the Yb II spectrum concern mostly the low-lying levels. Accurate cascade-free experimental measurements have been reported by Lowe *et al* (1993), Li *et al* (1999a), using laser-induced fluorescence, and by Berends *et al* (1993) and

Pinnington *et al* (1994, 1997), using a beam-laser technique. Electric-dipole amplitudes, lifetimes and polarizabilities of the low-lying levels of atomic ytterbium have been also considered by Porsev *et al* (1999). A compilation of the lifetimes available up to 1992 has been published by Blagoev and Komarovskii (1994).

The Rydberg states of Yb⁺ have been the subject of recent, but very limited, investigations. More specifically, the abnormal branching ratios in laser-excited series of this ion have been investigated by Zhao *et al* (1996) and by Xu *et al* (1997). Laser lifetime measurements, however, for highly excited levels along the series, are missing so far.

Data on the Yb II spectrum are needed in astrophysics, because the Yb⁺ ion has been observed in the sun (see e.g. Grevesse and Blanquet 1969, Gorshkov and Komarovskii 1986) and in chemically peculiar stars (see e.g. Cowley and Adelman 1983, Cowley 1984). The lack of reliable atomic data for many transitions observed, e.g. with the Hubble space telescope, has stimulated an extensive theoretical work by Fawcett and Wilson (1991). Additional theoretical efforts have been reported in different papers (Migdalek 1982, Biémont *et al* 1998, Li *et al* 1999a). In a recent paper, Migdalek and Siegel (2000) have addressed the difficult problem of the collapse of d and f orbitals along the isoelectronic sequence of Yb⁺.

In view of the complexity of the configurations involved in lanthanides, theoretical calculations need, however, whenever possible, to be tested by accurate experimental measurements. In particular, the discrepancies observed between experiment (Li *et al* 1999a, Yu and Maleki 2000) and theory (Fawcett and Wilson 1991) for some specific states call for additional theoretical and experimental investigations.

The purpose of the present paper is to provide new experimental laser lifetime measurements for selected levels along the Rydberg series of Yb⁺ for which no measurements are available. They extend the measurements reported recently by Li *et al* (1999a), using time-resolved laser-induced fluorescence spectroscopy. Comparisons with pseudo-relativistic Hartree–Fock (HFR) calculations, taking configuration interaction and core-polarization (CP) effects into account, are also reported in the present paper. As a consequence, the limits and the accuracy of the theoretical calculations for higher excitation levels, not yet considered experimentally, are more clearly assessed.

2. The calculations

The radiative lifetimes, τ_i , of the Yb II Rydberg states were calculated, using the well known expression

$$\tau_i = \frac{1}{\sum_j A_{ij}}$$

where A_{ij} represent the Einstein coefficients corresponding to all the possible radiative transitions depopulating the level i of interest. The method adopted for the calculations of these parameters is the so-called HFR approach described by Cowan (1981). In fact, for high Z (i.e. for all the lanthanide atoms and ions), it becomes desirable to include, in addition to the correlation effects, the most important relativistic corrections (mass–velocity effects and Darwin operator) in the non-relativistic differential equations in order to obtain the required radial wavefunctions. This can be done by considering a Pauli-type approximation to the Dirac–Hartree–Fock equations (see Cowan (1981) for more details).

The theoretical approach followed here is thus similar to that adopted in our recent papers concerning the same ion (Biémont *et al* 1998, Li *et al* 1999a) and the details will not be repeated here. In particular, CP effects, which are expected to play an important role in the

singly ionized heavy element Yb^+ , were introduced in the calculations in the way described previously (see e.g. Biémont *et al* 1998). The static dipole polarizability of Yb III was that computed by Fraga *et al* (1976), i.e. $\alpha_d = 7.35 a_0^3$, where a_0 is the value of the first Bohr orbit of the hydrogen atom. The cut-off radius r_c was chosen equal to $1.462 a_0$. These parameter values are close to those adopted by Migdalek and Siegel (2000), i.e. $\alpha_d = 6.39 a_0^3$ and $r_c = 1.492 a_0$. The configurations used in the calculations are similar to those previously used (Biémont *et al* 1998), but, as we were interested in Rydberg states, the configuration sets were extended to higher members of the series (in fact up to $n = 13$). The configurations considered were finally $4f^{14}ns$ ($n = 6-13$) + $4f^{14}nd$ ($n = 5-13$) + $4f^{13}5d6p$ + $4f^{13}6s6p$ + $4f^{13}6p6d$ and $4f^{14}np$ ($n = 6-13$) + $4f^{14}nf$ ($n = 5-13$) + $4f^{13}6s^2$ + $4f^{13}6p^2$ + $4f^{13}5d^2$ + $4f^{13}6d^2$ + $4f^{13}6s6d$ + $4f^{13}5d6s$ + $4f^{13}5d6d$. In view of the computer limitations, it was decided not to include in the final step the five configurations with an open $5p$ shell which were considered in our previous calculation (Biémont *et al* 1998). Their effect on the final oscillator strengths is expected to be small and is partly taken into account by the scaling down of the Slater integrals and by the consideration of the CP model potential.

In the present work, the parameters deduced from the least-squares optimization procedure minimizing the discrepancies between observed and calculated energy levels and described previously (Biémont *et al* 1998) have been adopted for the low-lying configurations. The scaling factors of the F^k , G^k and R^k integrals were chosen to be equal to 0.85 while the spin-orbit parameters were left at their *ab initio* values. The parameters describing the additional configurations with higher energies were obtained from a new fitting procedure. The experimental energy levels were taken from the NIST compilation (Martin *et al* 1978). In fact, 342 energy levels are known experimentally, belonging to the configurations $4f^{14}ns$ ($n \leq 10$), $4f^{13}6s^2$, $4f^{14}nd$ ($n \leq 11$), $4f^{13}5d6s$, $4f^{14}np$ ($n \leq 7$), $4f^{13}6s6p$, $4f^{13}5d^2$, $4f^{13}5d6p$, $4f^{14}nf$ ($n \leq 14$) and $4f^{14}ng$ ($n \leq 6$).

The theoretical HFR lifetime values obtained in the present work are summarized in table 1 (column 4), where we also report the theoretical values calculated up to $n = 8$ for levels which are known experimentally. We also quote, in the same table, the theoretical results obtained previously (Biémont *et al* 1998).

3. The measurements

In the present work, the radiative lifetimes of five even-parity levels of Yb II have been measured, at the Lund Laser Centre (Lund, Sweden), by time-resolved laser spectroscopy (for a detailed description of the experimental set-up, see e.g. Li *et al* 1999a, 2001). These levels are $8s^2S_{1/2}$, $6d^2D_{3/2}$, $6d^2D_{5/2}$, $7d^2D_{3/2}$ and $7d^2D_{5/2}$. A two-step excitation scheme was used to populate these levels, in which the first excitation step is from the ground state $6s^2S_{1/2}$ to the $6p^2P_{3/2}^o$ state ($30\,392.23 \text{ cm}^{-1}$) and the second step is from $6p^2P_{3/2}^o$ to the studied level. The relevant information on the levels considered, the excitation transitions and the observed wavelengths is presented in table 2.

Two Q-switched Nd:YAG 532 nm lasers were used in our experiment. The pulse widths of both lasers are 8 ns. One of the Nd:YAG lasers was pumping a dye laser operating with DCM dye and then, by frequency-doubling in a KDP crystal, it was used to produce a beam of 328.937 nm light for the first excitation step. The second Nd:YAG laser beam was compressed to about 1.5 ns by a SBS compressor (Li *et al* 1999b). This beam of shorter pulses pumped another DCM dye laser for the second excitation step. To obtain the desired UV radiation, we used KDP and BBO crystals, and Raman shifting techniques (Li *et al* 2000). A laser-induced plasma, produced by an ablation laser, was used as an ion source. The ablation and excitation pulses were synchronized by external triggering from the same delay generator. Observed

Table 1. Radiative lifetimes (in ns) for $4f^{14}nl$ ($nl = 7-8s, 6-8d, 6-7p, 5-8f$) Rydberg levels in Yb II. Both the HFR results and the experimental results obtained in the present work are shown.

Configuration	J	E (cm ⁻¹)	This work		Previous
			HFR	Expt ^b	
7s	1/2	54 304	4.91	3.9 ± 0.3 ^c	4.60 ^a , 4.9 ^k , 35 ± 3 ^f
8s	1/2	73 040	7.65	5.1 ± 0.5	
6d	3/2	62 174	3.12	3.1 ± 0.3	
	5/2	62 559	4.04	3.6 ± 0.5	4.04 ^a , 4.7 ^k , 4.9 ± 0.2 ^f
7d	3/2	76 517	5.28	5.3 ± 0.6	
	5/2	76 676	3.40	4.1 ± 0.4	
8d	3/2	83 840	5.08		
	5/2	84 016	5.95		
6p	1/2	27 062	7.88		8.60 ^a , 7.0 ⁱ , 8.8 ^k , 8.9 ± 0.5 ^e , 8.10 ± 0.13 ^d , 7.1 ± 0.4 ^f , 6.9 ± 0.6 ^g , 8.0 ± 0.2 ^h
	3/2	30 392	6.61		7.23 ^a , 8.4 ^k , 5.0 ⁱ , 7.3 ± 0.6 ^e , 7.2 ± 0.8 ^g , 5.5 ± 0.3 ^f , 6.3 ± 0.3 ^h , 6.1 ± 0.6 ⁱ
7p	1/2	63 706	29.40		
	3/2	65 594	37.52		
5f	5/2	70 502	3.75		
	7/2	70 580	4.30		
6f	5/2	80 459	5.87		
	7/2	80 472	6.30		
7f	5/2	85 898	9.34		
	7/2	85 906	9.96		
8f	5/2	89 176	11.98		
	7/2	89 185	15.04		

^a Biémont *et al* (1998) (HFR + CP method).^b This work, except when otherwise indicated.^c Li *et al* (1999a) (time-resolved laser-induced fluorescence).^d Berends *et al* (1993) (laser excitation on a fast ion beam).^e Burshtein *et al* (1975) (delayed-coincidence measurements).^f Blagoev *et al* (1979) (delayed-coincidence method).^g Andersen *et al* (1975) (beam-foil spectroscopy).^h Lowe *et al* (1993) (laser-induced fluorescence technique).ⁱ Rambow and Shearer (1978) (Hanle effect). Result corrected for hyperfine structure effects (cf Lowe *et al* 1993).^j Lowe *et al* (1993) (many-body perturbation theory).^k Fawcett and Wilson (1991) (semi-empirical HFR method).**Table 2.** Levels measured and excitation schemes.

Level	Excited level ^a (cm ⁻¹)	Excitation wavelength in air (nm)	Observed fluorescence (nm)
6d ² D _{3/2}	62 174.10	314.554	282
6d ² D _{5/2}	62 559.02	310.789	296
8s ² S _{1/2}	73 039.61	234.409	217
7d ² D _{3/2}	76 517.21	216.734	299
7d ² D _{5/2}	76 676.31	215.989	511

^a Martin *et al* (1978).

fluorescence was collected by a fused-silica lens, filtered by a monochromator and finally detected by a Hamamatsu 1564U microchannel plate (MCP) photomultiplier tube. Transient signals were acquired by a Tektronix TDS 602 oscilloscope. The detector risetime is 0.2 ns, and the oscilloscope has a 1 GHz bandwidth and a 2 gigasamples/s sampling rate. The second excitation laser pulse needed also to be recorded for the convolutional fitting of the fluorescence curve in order to evaluate the lifetime value. For each level studied, a series of measurements, under different conditions, was performed to eliminate the possibility of systematic errors. More precisely, the delay time, the intensity of the excitation laser and also that of the ablation laser were modified during the experiments. This resulted in a change of the fluorescence signal magnitude. However, it was found that the lifetime values remained constant (within the statistical scattering). In addition, for most measurements of each level, a homogeneous magnetic field was added in the region of the ion source to optimize the detection of the signal (Zhang *et al* 2001).

The five experimental lifetimes measured in this work are reported in the fifth column of table 1, where they are compared to the theoretical HFR results.

4. Results and discussion

For the $4f^{14}nd$ ($n = 6-7$) levels, the experimental lifetimes obtained in the present work agree well (the differences reaching 0.6, 12.2, 0.4 and 17.1%, respectively) with the theoretical predictions, the largest discrepancy being observed for the $7d^2D_{5/2}$ level. The theory also predicts rather well the large difference between the two fine-structure levels for $n = 7$. Looking at the details, it appears that a stronger mixing occurs for $J = 5/2$ (the purity of the level being 59%, while it does reach 92% for $J = 3/2$). Consequently, the corresponding lifetime is more sensitive to the eigenvector composition and could eventually be less accurately determined. A similar behaviour is also observed, to a lesser extent, along the np series, where the eigenvector purities are found to be equal to 85% ($6p_{1/2}$), 70% ($6p_{3/2}$), 79% ($7p_{1/2}$) and 56% ($7p_{3/2}$). On the other hand, the purities of the $nf_{5/2,7/2}$ ($n = 6-8$) levels are nearly constant ($\sim 99\%$), which explains the very small variation of the J -lifetimes along the nf Rydberg series. For the ns levels, the HFR values appear systematically larger than the experimental measurements, the new result for $n = 7$ even increasing the difference slightly. For $7s^2S_{1/2}$, the agreement with the previous theoretical result (Biémont *et al* 1998) remains reasonable, the discrepancy between theory and experiment reaching 25%. There is no clear explanation for the larger difference (50%) appearing for 8s. It is reasonable to consider that the HFR values are possibly less accurate for larger values of n than for the low excitation states, because many levels belonging to the higher configurations ($4f^{13}5d6p$, $4f^{13}6p6d$, ...) might possibly play an increasing role in the configuration mixings. This hypothesis is confirmed by the fact that the dominant eigenvector components are rather different for the two 8d levels (61 and 77%, respectively) and differently sensitive to the configuration set adopted. It is also observed that the ratio between the theoretical values for 7s and 8s (i.e. 1.31) is somewhat different from the experimental result (i.e. 1.56). Much larger configuration sets would probably be necessary to definitely settle this question, but would require a substantial computational effort combined with laboratory analyses for determining additional levels.

We also present in table 1 our theoretical HFR results for the odd parity levels up to $n = 8$. Many measurements are available for the two $6p^2P^o$ levels, but they are characterized by a rather large scatter. Our HFR result for $6p^2P^o_{1/2}$ is in close agreement with the laser measurements of Berends *et al* (1993) and of Lowe *et al* (1993), which are expected to be accurate. For $6p^2P^o_{3/2}$, our theoretical lifetime agrees well with the value published by Lowe *et al* (1993), with the Hanle effect measurement by Rambow and Shearer (1978) corrected for

hyperfine structure effects, and it is within the error bars affecting the beam–foil measurement of Andersen *et al* (1975). No results are available for comparison for nf ($n \leq 8$) $^2F^o$ states.

An independent test of the present theoretical model is additionally provided by the consideration of the lifetimes of the $4f^{14}5d^2D_{3/2}$ and $^2D_{5/2}$ levels. The $4f^{14}5d^2D_{3/2}$ metastable level can decay to the $4f^{14}6s^2S_{1/2}$ ground state through M1 and E2 transitions, the M1 contribution appearing, however, negligibly small when compared to the E2 one. These lifetimes have been measured using a technique based on the consideration of ion clouds in buffer gases (Gerz *et al* 1988), but this method can lead to considerable errors, particularly for long-lived states (Yu *et al* 1997). More recent measurements are due to Yu and Maleki (2000) using single ytterbium ions confined in an rf trap. This technique has the advantage of eliminating all possible pressure effects. The experimental lifetimes of Yu and Maleki (2000), i.e. $\tau = 52.7 \pm 2.4$ and 7.0 ± 0.4 ms for $^2D_{3/2}$ and $^2D_{5/2}$ levels, respectively, agree well with previous experimental data, i.e. $\tau = 52 \pm 1$ ms (Gerz *et al* 1988) and $\tau = 7.2 \pm 0.3$ ms (Taylor *et al* 1997). These authors emphasize the discrepancies between their results and theoretical estimates (Fawcett and Wilson 1991). Another theoretical value, however, not considered by Yu and Maleki (2000), obtained for $^2D_{5/2}$ (Biémont and Quinet 1998), i.e. $\tau = 51.8$ ns, was in agreement with their measurement. It is interesting to compare the results obtained using the present theoretical model with the accurate measurements of Yu and Maleki (2000). They are $\tau = 52.8$ and 6.9 ms, respectively, and appear in excellent agreement with the experiment.

5. Conclusions

New radiative lifetimes have been calculated and also measured by time-resolved laser-induced fluorescence spectroscopy for Rydberg states of Yb II. The A -values of the transitions depopulating the levels of interest will be useful for analysing the spectra of chemically peculiar stars, because the line identification and profile analysis in astrophysical spectra have been prevented up to now mostly by the lack of relevant radiative parameters. The excitation energies of the depopulating channels of the levels measured are comparable to those of the transitions recently observed in the visible spectral range of astrophysical spectra like those of the Przybylski star (Cowley *et al* 2000) and, consequently, these transitions are expected to appear in stellar spectra. The present paper provides also useful information for assessing the theoretical methods usable for atomic processes calculations in this metrologically important ion. The new theoretical lifetimes obtained for the nf series also call for additional experimental work on this ion.

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