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Experimental and theoretical lifetimes in Yb III

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Abstract. Lifetimes of three levels belonging to the configuration $4f^{13}5d$ with $J = 1$ in Yb III have been measured for the first time using the time-resolved laser-induced fluorescence method. Experimental transition probabilities have been deduced for the transitions between the levels studied and the ground state. The comparison of the experimental lifetimes with theoretical data, deduced within the relativistic Hartree-Fock (HFR) approach, underlines the dramatic importance of an adequate consideration of core-polarization effects in the theoretical model and the sensitivity of one of the lifetime values to small correlation effects.

PACS. 32.70.Cs Oscillator strengths, lifetimes, transition moments – 42.62.Fi Laser spectroscopy

1 Introduction

The interest in spectral data of rare-earth (RE) elements or ions is due primarily to their importance in astrophysics in relation with their observation in the spectra of chemically peculiar stars. Many laboratory analyses have been devoted to the investigation of neutral or singly ionized lanthanide atoms [1]. A number of spectral lines of doubly-ionized RE atoms have been identified in stellar spectra [2–6] but the line identifications and abundance determinations frequently suffer from the lack of reliable atomic data. Consequently, additional laboratory data regarding the intensities of the lines in the third spectra of lanthanides are needed.

In the last few years, term analyses and transition probability determinations have been performed for the third spectra of some RE elements, such as Dy III [7], Er III [8,9], Ce III [10,11], Nd III [12], La III and Lu III [13], Pr III [14,15], Tm III [16] and Yb III [17]. These analyses have motivated laser lifetime measurements for the excited levels of doubly-ionized RE atoms, such as La III [18], La III and Lu III [13], Eu III [19], Lu III [20,21], Ce III [22], Tm III [16], Pr III [15], Er III [9], Gd III [23] and Yb III [17], in order to obtain reliable experimental data to test the theoretical models. In view of the "complexity" of the configurations involved, containing an open $4f$ shell, these models are strongly dependent upon the computer capabilities available and only a limited amount of correlation can be introduced explicitly in the calculations. This makes the theoretical models dependent upon semi-empirical parametrization in an attempt to mimic the interactions with the myriad of distant configurations not considered explicitly in the configuration interaction process. This parametrization can be made *via* the consideration of core-polarization (CP) contributions but this approach in turn is partly hampered by the well-known fast collapse of $4f$ orbital in lanthanides starting at lanthanum [24].

In Yb III, two lifetimes of levels within the configuration $4f^{13}6p$ have been previously measured using time-resolved laser-induced fluorescence [17]. In the same paper, theoretical calculations had also been performed with the HFR approach including CP effects and the oscillator strengths of the transitions between the measured levels and those of the configurations $4f^{13}5d$ or $4f^{13}6s$ had been deduced from a combination of the experimental lifetime values with the relevant theoretical branching fractions.

In the present paper, the lifetimes of three additional $4f^{13}5d$ levels with $J = 1$ in Yb III have been measured at the Lund Laser Centre (LLC) with the time-resolved laser-induced fluorescence method and the associated CP effects are analyzed theoretically. In the measurements, the excitation laser has been focused to avoid the influence of flight-out-of-view effects on long lifetime measurements and a suitable magnetic field has been applied over the plasma produced by laser ablation in order to reduce the background plasma light.

2 Experimental set-up

The three Yb III levels studied in the present paper can be excited from the ground state, and each of them has only one decay channel. In order to measure their

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Table 1. Measured levels and excitation schemes.

Level[a]		Energy[a]	Excitation			λ_{obs}
			Dye	λ_{exc}	Conversion	
jj	*LS*	(cm^{-1})	used	$(nm)_{vac}$	scheme[b]	$(nm)_{vac}$
$(7/2, 5/2)^0_1$	$^3P_1$	39 720.79	DCM	251.757	$3\nu + 2S$	251.757
$(5/2, 5/2)^0_1$	$^3D_1$	50 029.42	R610	199.882	3ν	199.882
$(5/2, 3/2)^0_1$	$^1P_1$	53 365.19	R640	187.388	$3\nu + AS$	187.388

[a] From the NIST database at the address: `http://www.physics.nist.gov/cgi-bin/AtData/`
[b] 3ν: frequency tripling; 2S: second Stokes component; AS: anti-Stokes component.

Table 2. Experimental and theoretical radiative lifetimes (in ns) in Yb III.

Level	Energy (cm^{-1})	$\tau_{exp.}$	τ_{theory}	
			A	B
$4f^{13}(^2F^0_{7/2})5d_{5/2}(7/2, 5/2)^0_1$	39 720.79	230 (20)	166	270
$4f^{13}(^2F^0_{5/2})5d_{5/2}(5/2, 5/2)^0_1$	50 029.42	280 (30)	272	476
$4f^{13}(^2F^0_{5/2})5d_{3/2}(5/2, 3/2)^0_1$	53 365.19	11.4 (7)	6.3	10.3

Theory: A for calculation A, B for calculation B (see the text).

lifetimes, different dyes and different nonlinear conversions are needed. Measured levels, employed dyes and excitation schemes are shown in Table 1.

The experimental set-up can be seen in Figure 1. Free Yb^{2+} ions were obtained from a laser-induced plasma. Laser pulses, characterized by a 532 nm wavelength, a 10 Hz repetition rate and 10 ns duration, emitted from a Nd:YAG laser (Continuum Surelite) with 0–50 mJ tunable pulse energy, were focused vertically onto the surface of an Yb foil rotated in a vacuum chamber. After the laser pulse, the plasma expands from the foil. In the plasma, atoms and ions in different ionization stages, are moving at different speeds. In order to obtain the required excitation laser light, 8 ns pulses from another Nd:YAG laser (Continuum NY-82 injecting seeded) were sent to a Stimulated Brillouin Scattering (SBS) compressor to shorten the pulses. The pulse duration of the output from the SBS compressor was about 1 ns. A short excitation pulse was needed since one of the levels studied was short-lived. The laser was used to pump a dye laser (Continuum Nd-60). DCM, R610 and R640 dyes were used separately in the lifetime measurements for the different levels. The dye laser was frequency-tripled in a nonlinear optical system which consisted of a KDP crystal, a retarding plate and a BBO crystal. The third harmonic from the system was focused by a quartz lens into a hydrogen cell with 10 bars pressure to produce the desired wavelength by stimulated Raman Scattering. The excitation laser light needed in the measurements was selected using a CaF_2 Pellin-Broca prism and focused by a CaF_2 lens at the centre of the vacuum chamber, about 10 mm above the foil.

The two Nd:YAG lasers were triggered externally by a digital delay generator (Stanford Research System Model 535), used to adjust the delay time between the ablation and excitation pulses. After the ablation pulse, the ions were excited by the excitation-laser beam, which passed through the plasma horizontally. The decay fluorescence from the level measured was imaged by two CaF_2 lenses onto the entrance slit of a vacuum monochromator, and detected with a Hamamatsu R3809U-58 photomultiplier. The time-resolved signal was acquired and averaged using a digital transient recorder (Tektronix Model DSA 602). The averaged time-resolved fluorescence decay curve was sent to a personal computer for lifetime evaluation. The experimental lifetime values are reported in Table 2.

3 Measurements and results

Since there was only one decay channel for each level measured, it was verified carefully that the correct level was excited. Firstly, all the identified lines of Yb I and Yb II were checked in order to make sure that only the Yb III lines of interest appeared in the spectral neighbourhood of the excitation wavelength. Furthermore, the modification of the fluorescence signal as a function of the delay times was considered. Commonly, the speed of Yb III ions is larger than that of Yb II and Yb I ions, and this is expected to be reflected by the occurrence of a maximum fluorescence signal at shorter delay times.

The entrance slit of the monochromator was placed vertically to avoid possible flight-out-of-view effects due to long lifetimes for the two levels at 39 720.79 cm^{-1} and at 50 029.42 cm^{-1}. Possible influence of such effects was evaluated and found negligible. During the lifetime data registration, the delay time was varied within the 1.6–2.0 μs range. Usually, the fluorescence decay within a time span interval covering more than 4 lifetimes duration needs to be recorded in order to get a decay curve suitable for accurate lifetime determinations. For the delay time of 2.0 μs, corresponding to measurements on ions with a speed of about 5 km/s, the required slit height, which should be

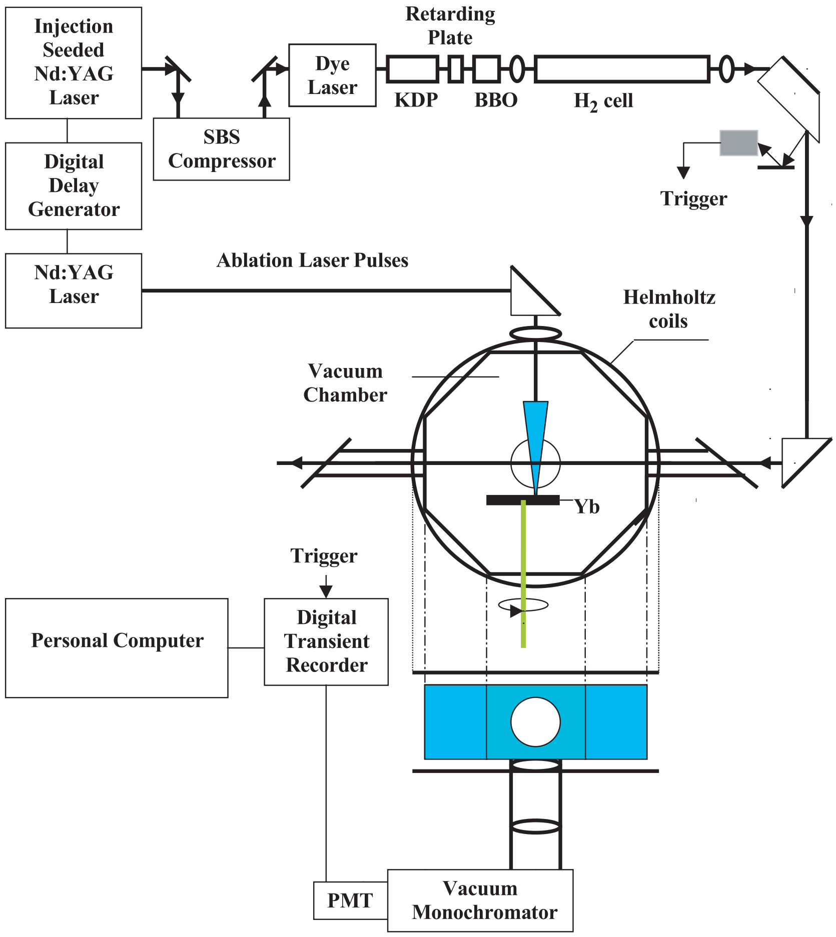

Fig. 1. Experimental set-up used in the lifetime measurements.

larger than 7 mm, reached in fact 12 mm in our experiments.

As a further refinement, a suitable magnetic field of about 60 Gauss, was added in the plasma zone by a pair of Helmholtz coils. This field made the ions in different ionization stages and, consequently, characterized by different speeds, run in different traces. The consequence was not only the elimination of the possible quantum beat effects, but also the weakening of the background associated with the ablation laser and the plasma recombination processes.

During the experiments, a variety of experimental conditions were modified, including the delay time, the intensity of the excitation laser and also that of the ablation laser. This resulted in a change of the fluorescence signal magnitude by a factor of 10. However, it was found that the lifetime values remained constant. In order to obtain a reasonable signal-to-noise ratio, a fluorescence decay curve was obtained by averaging fluorescence photons from more than 2 000 pulses. A typical curve is seen in Figure 2. The lifetime evaluation was performed by an exponential fit and, for each level, the lifetime value was obtained through the averaging of more than 20 curves. The three lifetimes measured are shown in Table 2, where the quoted uncertainties reflect not only the statistical errors, but also possible small remaining systematic errors.

Because each measured level has only one decay channel, the transition probabilities and oscillator strengths of the lines originating from the levels studied to the ground state were deduced directly from the lifetime data measurements. They are given in Table 3.

4 Theoretical calculations

In RE ions, previous calculations of transition probabilities [17,25] for Yb II and Yb III, respectively, have emphasized the importance of CP effects in relation with the limited sets of configurations one is able to consider explicitly in Cowan's HFR code [24]. The current CP models [26] are generally able to reproduce, with an excellent accuracy, the results deduced from the laser measurements for the nl-$n'l'$-type transitions with nl, $n'l' \neq 4f$ but the f values of the $4f$-nd transitions, calculated with the same model, differ sometimes substantially from the experimental results. This statement imposes the application of a semiempirical scaling factor to the $\langle 4f|r|5d\rangle$ dipole operator, this procedure allowing to compensate for the sudden collapse of the $4f$ orbital largely embedded in the $5s$ and

Table 3. Experimental transition probabilities (10^8 s^{-1}) and oscillator strengths in Yb III.

Upper level	λ (nm)	gA	Log(gf)
$4f^{13}(^2\mathrm{F}^0_{7/2})5d_{5/2}(7/2,5/2)^0_1$	251.76	0.13 (1)	–1.91
$4f^{13}(^2\mathrm{F}^0_{5/2})5d_{5/2}(5/2,5/2)^0_1$	199.88	0.11 (1)	–2.19
$4f^{13}(^2\mathrm{F}^0_{5/2})5d_{3/2}(5/2,3/2)^0_1$	187.39	2.63 (6)	–0.86

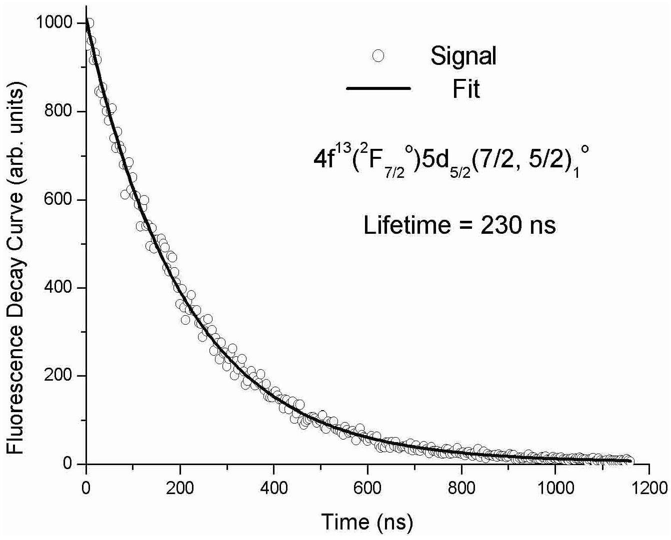

Fig. 2. A typical experimental time-resolved fluorescence signal from the level at 39 720.79 cm^{-1} in Yb III. The recorded data points are marked as o in the figure. The solid curve shows the exponential fit curve for the LIF signal. The lifetime deduced from the fit was 230 ns.

$5p$ core orbitals, this collapse being not taken properly into account by the model. In an attempt to evaluate quantitatively the importance of these effects in Yb III, several different calculations were performed in the present work.

In our previous work on Yb III (calculation A) [17], intravalence correlation was considered among the configurations $4f^{14}$, $4f^{13}np$ ($n = 6$–7), $4f^{13}nf$ ($n = 5$–7) and $4f^{13}nd$ ($n = 5$–7), $4f^{13}ns$ ($n = 6$–7) for even and odd parities, respectively. The CP effects were then included in the model using the CP potential and the corrected dipole moment of the transition deduced from the work of Migdalek and Baylis [26] with a dipole polarisability of the ionic core $\alpha_\mathrm{d} = 5.40a_0^3$ and a cut-off radius $r_\mathrm{c} = 1.40a_0$. These polarisation corrections were neither introduced in the atomic orbital calculations of the ground configuration, $4f^{14}$, nor in the $\langle 4f|r|5d\rangle$ dipole operator. Instead, a scaling factor (SF), equal to 0.82, was applied to this matrix element. This SF was deduced from a curve showing the ratio between CP corrected and uncorrected transition matrix elements for transitions not involving a $4f$ electron as a function of the uncorrected matrix element. The lifetime values obtained using this approach for the three levels of interest are listed in Table 2 under the heading A. It is seen that theory and experiment agree well for the level at 50 029.42 cm^{-1} while large discrepancies are observed for the other two levels.

In the present paper, a more extensive calculation was performed (calculation B) in which CP was considered by including explicitly in the physical model some configurations with excitation from the $5s$, $5p$ and $4f$ core-orbitals. More precisely, in addition to the configurations retained in calculation A, we did include $4f^{12}5d^2$, $4f^{12}6s^2$, $4f^{12}6p^2$, $4f^{12}5d6s$, $5s^25p^54f^{13}5d^2$, $5s5p^64f^{14}5d$ and $4f^{12}5d6p$, $4f^{12}6s6p$, $5s^25p^54f^{14}5d$, $5s5p^64f^{13}5d^2$ for even and odd parities, respectively. The effect of opening the $5s$ cadmium and $5p$ xenon cores was mimicked through the consideration of the configurations $5s^25p^54f^{13}5d^2$, $5s5p^64f^{14}5d$ and $5s^25p^54f^{14}5d$, $5s5p^64f^{13}5d^2$. The lifetime values, deduced in the framework of calculation B, are also reported in Table 2 (heading B). For the two levels at 39 720.79 and 53 365.19 cm^{-1}, the agreement theory-experiment is good. A larger discrepancy is observed for the level at 50 029.42 cm^{-1} for which the theoretical lifetime has been found very sensitive to small energy differences between the $^3\mathrm{D}^0_1$ and $^1\mathrm{P}^0_1$ levels (for the three $J = 1$ levels, LS coupling designations are more adequate than the jj coupling ones). In order to further investigate this problem, additional calculations were performed using the same set of configurations as in calculation B but considering slightly different numerical values of the $F^2(4f, 5d)$ Slater integral (from 19 000 cm^{-1} to 24 000 cm^{-1}), which resulted in a modification of the percentage composition of the $^3\mathrm{D}^0_1$ level with an admixture of the $^1\mathrm{P}^0_1$ percentage ranging from 1.6% to 4.1%. Then we found that the lifetime values for the 39 720.79 and 53 365.19 cm^{-1} levels were very stable (the differences did not exceed 9%) while the lifetime values for the level at 50 029.42 cm^{-1} differed by up to a factor of two. This indicates the sensitivity of the 50 029.42 cm^{-1} level lifetime to the mixing between the three $4f^{13}5d$ $J = 1$ levels of Yb III.

Furthermore, it was verified that the results of calculation B could be retrieved by using the smaller configuration set of calculation A, provided that an adequate SF was applied to the $\langle 4f|r|5d\rangle$ dipole operator. The best SF for the three levels was found to be equal to 0.62 (thus *lower* than the value of 0.82 adopted in calculation A) and the corresponding lifetimes (*i.e.* 273, 448 and 10.4 ns, respectively) were very close to the theoretical values obtained in the more extensive calculation B but with a persistent discrepancy for the level $(5/2, 5/2)^0_1$.

As a conclusion, the achievement of an agreement theory-experiment simultaneously for the three levels considered in the present work would probably necessitate to consider explicitly a substantially larger set of configurations in the calculations than that mentioned in calculation B in order to describe, in a more realistic way, the exact mixing occurring between the $J = 1$ low lying levels in Yb III. Such calculations were prevented in the present work basically by the computer capabilities available.

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