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Li, Z. S; Svanberg, Sune; Quinet, P; Tordoir, X; Biemont, E

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Lifetime measurements in Yb II with time-resolved laser spectroscopy

Z S Li†, S Svanberg†, P Quinet‡, X Tordoir§ and E Biémont‡§

† Department of Physics, Lund Institute of Technology, PO Box 118, S-221 00 Lund, Sweden
‡ Astrophysique et Spectroscopie, Université de Mons-Hainaut, B-7000 Mons, Belgium
§ Institute of Nuclear Experimental Physics (Bât. B 15), University of Liège, B-4000 Liège, Belgium

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Abstract. Natural radiative lifetimes of 10 levels of singly ionized ytterbium have been measured with time-resolved laser spectroscopy. Free Yb^+ ions were produced in a laser-induced plasma. A stimulated Brillouin scattering technique has been used to produce laser pulses as short as 1 ns for the excitation of short-lived Yb^+ states. Pseudo-relativistic Hartree–Fock calculations, taking into account the core-polarization effects, have also been performed. A good agreement of the measured and the calculated lifetime values has been achieved.

1. Introduction

The interest for astrophysics in the Yb II spectrum has stimulated a number of theoretical and experimental studies. Further, the Yb ion is much studied for atomic clock applications. For the lifetimes of the low-lying excited states, beam–foil measurements (Andersen *et al* 1975), delayed coincidence measurements (Burshtein *et al* 1974) and Hanle effect measurements (Rambow and Shearer 1976) have been performed. Blagoev *et al* (1978) measured a few lifetimes of some high-lying excited states using the delayed-coincidence method with a fast electron-beam excitation. Most of the lifetime values published before 1992 can be found in the compilation made by Blagoev and Komorovskii (1994).

A comprehensive theoretical work based on HFR calculations has been performed by Fawcett and Wilson (1991), who provided a large number of oscillator strengths and lifetime values for comparison with the experimental observations. Since then, precise lifetime measurements with cascade-free selective laser excitation, such as beam–laser (Berends *et al* 1993, Pinnington *et al* 1994, 1997) and laser-induced fluorescence (Lowe *et al* 1993), have become available.

In recent theoretical works (Biémont *et al* 1998, Biémont and Quinet 1998), improved HFR calculations with consideration of extended configuration interaction and core-polarization effects have been performed. The calculated lifetime values agreed very well with the existing experimental data, but a few levels which were measured by the delayed-coincidence method (Blagoev *et al* 1978) showed large deviations. The present work is devoted to lifetime measurements of these high-lying upper levels using selective laser excitation followed by a time-resolved observation, to check the reliability of the theoretical approach.

One- or two-step excitations have been used, as indicated in figure 1 and in table 1, to populate the desired upper levels. To enable us to make accurate measurements of the short

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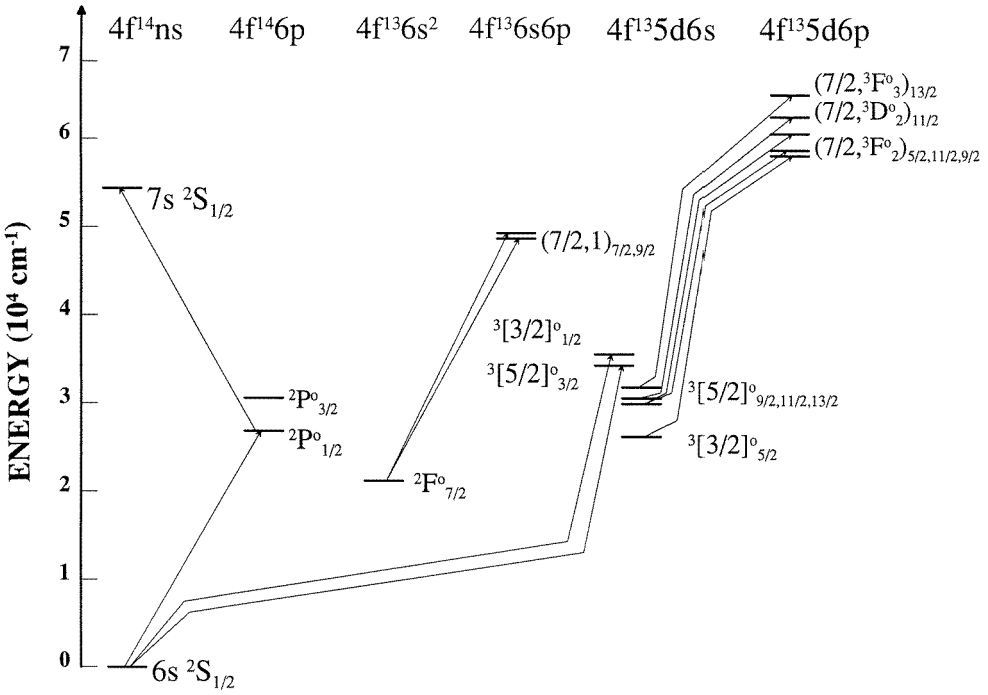

Figure 1. Partial energy level diagram of Yb^+ with relevant excitation pathways indicated.

Table 1. Levels measured and excitation scheme.

Level[a]	E_{exp}[b] (cm^{-1})	Excitation		Observed fluorescence (nm)
		Origin (cm^{-1})	λ (nm)	
$4f^{13}5d6s\ ^2F^o_{7/2}[^3D\ 5/2]^o_{3/2}$	32 981.59	0	303.12	303.1
$4f^{13}5d6s\ ^2F^o_{7/2}[^3D\ 3/2]^o_{1/2}$	33 653.86	0	297.06	297.1
$4f^{13}6s6p(^2F^o_{7/2}, {}^3P^o_1)_{7/2}$	48 900.41	21 418.75	363.78	451.5
$4f^{13}6s6p(^2F^o_{7/2}, {}^3P^o_1)_{9/2}$	49 301.16	21 418.75	358.55	533.5
$4f^{14}7s\ ^2S_{1/2}$	54 304.30	0\27 061.82	369.42\366.97	418.1
$4f^{13}5d6p(^2F^o_{7/2}, {}^3F^o_2)_{5/2}$	58 823.58	26 759.02	311.79	267.3
$4f^{13}5d6p(^2F^o_{7/2}, {}^3F^o_2)_{11/2}$	58 961.37	30 224.33	347.89	352.0
$4f^{13}5d6p(^2F^o_{7/2}, {}^3F^o_2)_{9/2}$	61 214.66	30 224.33	322.59	251.2
$4f^{13}5d6p(^2F^o_{7/2}, {}^3F^o_3)_{13/2}$	61 873.40	30 562.79	319.29	330.6
$4f^{13}5d6p(^2F^o_{7/2}, {}^3D^o_2)_{11/2}$	63 944.18	31 631.59	309.39	299.5

[a] Designation according to Fawcett and Wilson (1991).
[b] From Martin *et al* (1978).

lifetimes, the stimulated Brillouin scattering (SBS) phenomenon has been employed as part of the production of laser pulses of as short as 1 ns duration. Experimental details of this technique have been described by Li *et al* (1998).

2. Experimental set-up

The experimental set-up used in the present experiment is shown in figure 2. Two Nd:YAG lasers (Continuum NY-82) were used to pump a dye laser (Continuum ND-60) and a Ti:sapphire laser (TS-60), respectively. To produce short laser pulses, the 8 ns output of one of the Nd:YAG lasers was compressed in an SBS unit, before being used to pump the dye laser. Tunable laser pulses of about 1 ns duration were obtained from the dye laser. To cover the spectral region needed in this experiment, different red dyes (LDS 590, 610, 640 and 698) have been used in the dye laser. The output from the dye laser and the Ti:sapphire laser was frequency doubled in a KDP crystal. For the two-step excitation, the shorter laser pulses were used for the second step; for the single-step excitation, only the short laser pulses were employed. For both cases, the transient digitizer was triggered by the shorter laser pulses.

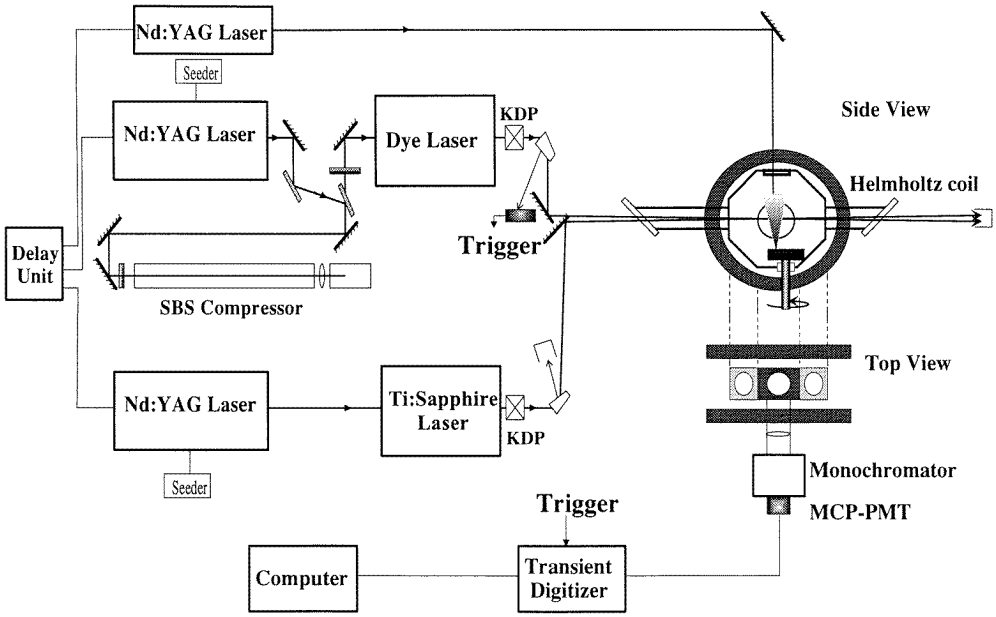

Figure 2. Experimental set-up.

Free Yb^+ ions were obtained from a laser-induced plasma. A separate Nd:YAG laser (Continuum Surelite) provided 532 nm, 10 ns pulses of roughly 10 mJ pulse energy, which were focused perpendicularly onto a rotated metallic ytterbium target in the vacuum chamber. The excitation laser beam was sent directly through the plasma about 10 mm above the ablation spot. The Nd:YAG lasers were synchronized by external triggering from a delay unit (Stanford Research Systems Model 535 digital delay generator), enabling a free variation of the delay time between the excitation pulses and the ablation pulses. Laser-induced fluorescence from the selected upper levels, collected by a fused-silica lens and appropriately filtered by a $\frac{1}{8}$ m monochromator (resolution 6.4 nm mm^{-1}), was detected by a Hamamatsu 1564u microchannel-plate (MCP) photomultiplier (200 ps rise time). The data acquisition was performed by a digital transient recorder (Tektronix Model DSA 602) which has a 1 GHz bandwidth and worked in real-time with a 2 GS s^{-1} sampling rate. The averaged time-resolved fluorescence decay curves were transferred to a personal computer and lifetime evaluations were performed in direct connection with the experiments.

3. Measurements and results

A partial energy level diagram of Yb^+ relevant to this experiment is shown in figure 1. Starting from the ground state $4f^{14}6s\ ^2S_{1/2}$, a two-step procedure was adopted to excite the $4f^{14}7s\ ^2S_{1/2}$ level. The frequency-doubled output at 369.42 nm from the Ti:sapphire laser was used to excite the $4f^{14}6p\ ^2P^o_{1/2}$ level in a first step, and the 366.97 nm frequency-doubled laser pulses from the dye laser further excited the ions to the desired upper level. Temporal and spatial overlaps of the two laser pulses were fine tuned to optimize the fluorescence signal. Single-step excitation was used to populate the other desired upper levels from the ground state or from a metastable state. A sufficient population of the metastable states can be obtained for appropriately chosen plasma conditions. To make sure that the right level was excited, all the possible decay channels were checked by tuning the monochromator, and then proper transitions were selected for recording the decay curves (see table 1).

The plasma density and temperature at the observed point can be adjusted by changing the ablation pulse energy, the size of the focus point, the distance above the target surface and the delay time between excitation and ablation pulses. For the measurements which started from the ground state, the delay time between the ablation beam and the excitation beam can be as long as 30 μs, while still obtaining a reasonably strong signal to evaluate the lifetime. The measured lifetimes in the delay time interval between 7 and 30 μs remained constant within the experimental scattering, while the detected fluorescence intensity varied by a factor of 20. For the levels excited from metastable states, the delay time could be varied only within 0.5–1.5 μs to keep a reasonably strong fluorescence signal, but still the detected intensity varied by a factor of 10, while the evaluated lifetime values remained constant. This indicated that the collision quenching and radiation trapping effects were negligible under our measurement conditions.

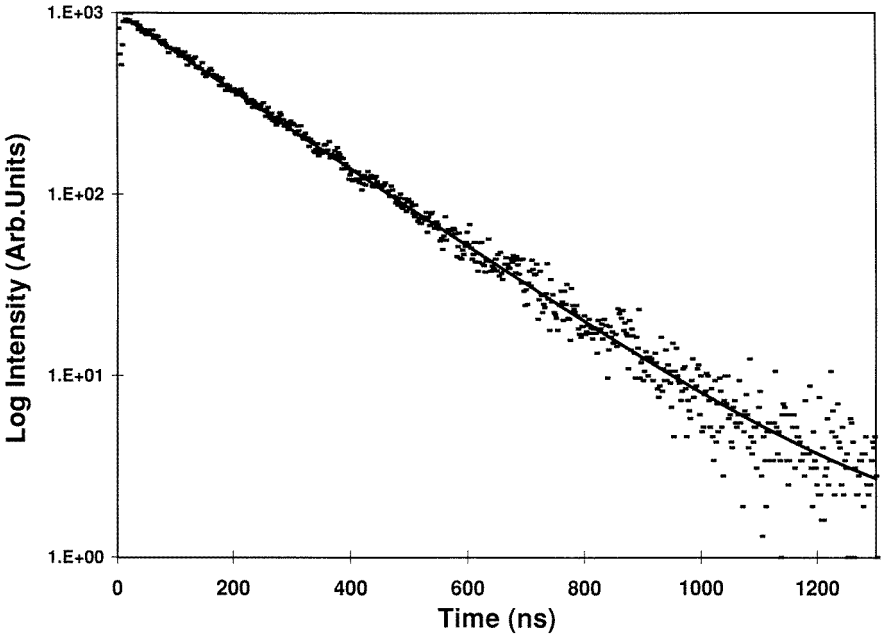

Figure 3. Detected time-resolved fluorescence signal from the $4f^{13}5d6s\ ^2F^o_{7/2}[^3D\ 5/2]^o_{3/2}$ state with an exponential fit which gives a lifetime of 196 ns.

The influence of possible hyperfine structure beats was checked for each level by setting the polarization direction of the excitation laser beam equal to 54.7°, 35.3° or 90° to the detection direction. No influence of the curve shape was found for the different settings (see Hannaford and Lowe (1983) for a detailed theoretical analysis). For the long-lived states, a sufficiently strong magnetic field, provided by Helmholtz coils, was utilized to wash out Zeeman quantum beats. Possible flight-out-of-view effects were specially investigated by changing the monochromator slit width and position. The good exponential fit of the detected signal as shown in figure 3 proved a full elimination of the flight-out-of-view effect. For the long-lived states, a pure exponential fit was performed to obtain the lifetime values, while for the states which had a lifetime shorter than 5 ns, a fit to the detected curve of a convolution of the excitation pulse and an exponential was performed. The temporal shape of the excitation pulse was recorded with the same detection system by detecting the directly scattering light of the laser pulse from a metal rod which was inserted into the interaction spot of the Yb^+ beam and the excitation beam. To ensure the precision of this convolution fitting, saturation of the excitation was effectively prevented by inserting neutral-density filters in the excitation laser beam to reduce the power. A typical decay curve and the corresponding convolution fit are shown in figure 4.

To ensure a linear response of the detection system, only sufficiently weak signals were detected for each pulse in the measurements. An average of 1000–4000 pulses was necessary for each curve depending on the signal-to-noise ratio. Around 30 curves for each level have been recorded and the average lifetime value was adopted as the final results. One standard deviation in the data and estimated possible systematic errors have been introduced in the error bars. The experimental lifetime results are given in table 2.

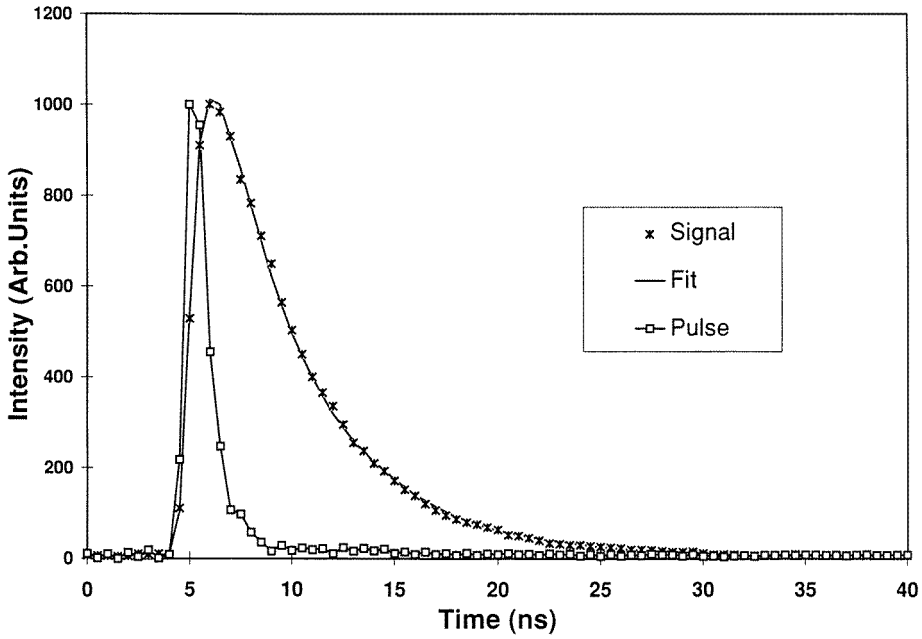

Figure 4. Detected time-resolved fluorescence signal from the $4f^{14}7s\ ^2S_{1/2}$ state and the recorded excitation laser pulse. The convolution procedure gives a lifetime of 3.9 ns.

Table 2. Observed and calculated lifetimes and comparison with previous results.

	Lifetime value (ns)			
	This work		Previous work	
E_{exp}[b] (cm^{-1})	Expt	HFR	Expt	Calc.
32 981.59	196(20)	157.4[a]	16(1)[c]	137.2[f]
33 653.86	39(3)	32.10[a]	37.7(5)[d], 42(3)[c]	17.2[f]
48 900.41	33(3)	31.8		
49 301.16	30(3)	29.1		
54 304.30	3.9(3)	4.60[a]	35(3)[c]	4.9[f]
58 823.58	3.8(3)	3.73[a]	3.59(7)[e]	3.1[f]
58 961.37	6.1(4)	4.87[a]	15(1)[c]	4.1[f]
61 214.66	4.7(4)	3.96		
61 873.40	5.1(4)	4.05		
63 944.18	3.8(3)	3.15		

[a] Biémont *et al* (1998) (HFR method, see the text).
[b] From Martin *et al* (1978).
[c] Blagoev *et al* (1978) (delayed coincidence).
[d] Berends *et al* (1993) (beam–laser measurements).
[e] Pinnington *et al* (1994) (beam–laser measurements).
[f] Fawcett and Wilson (1991) (HFR method).

4. HFR calculations

A relativistic Hartree–Fock calculation, taking configuration interaction and polarization effects into account, has been combined with a least-squares optimization procedure of the Slater and spin–orbit integrals. As the method has been described elsewhere (Biémont *et al* 1998), only a short summary is presented here. Configuration interaction has been considered among the configurations $4f^{14}ns$ ($n = 6$–7) + $4f^{14}nd$ ($n = 5$–7) + $4f^{13}5d6p$ + $4f^{13}6s6p$ + $4f^{13}6p6d$ + $5p^54f^{14}5d6p$ + $5p^54f^{14}6s6p$ and $4f^{14}np$ ($n = 6$–7) + $4f^{13}6s^2$ + $4f^{13}6p^2$ +$4f^{13}nd^2$ ($n = 5$–6) + $4f^{13}6s6d$ + $4f^{13}5d6s$ + $4f^{13}5d6d$ + $5p^54f^{14}5d^2$ + $5p^54f^{14}6s^2$ + $5p^54f^{14}5d6s$. The core-polarization effects were considered using the expressions of the core-polarization potential and of the corrected dipole moment of the transition deduced from the work of Migdalek and Baylis (1978). The dipole polarizability of the ionic core, α_d, and the cut-off radius, r_c, were chosen equal to $\alpha_d = 7.35\alpha_0^3$ (dipole polarizability of Yb III as computed by Fraga *et al* (1976)) and to $r_c = 1.462\alpha_0$ (the HFR average value $\langle r \rangle$ of the outermost core orbital of the investigated valence configuration, i.e. $5p^6$). The HFR radiative lifetimes have been found to agree very well with the relativistic quantum defect orbital (RQDO) theory and also with the most accurate laser measurements (see Biémont *et al* 1998, Biémont and Quinet 1998 for a discussion). The calculated HFR lifetime values are compared with the laser measurements in table 2.

5. Discussion

The measurements of this experiment agree very well with the existing beam–laser results (Berends *et al* 1993, Pinnington *et al* 1994) for the $4f^{13}5d6s\ {}^2F^o_{7/2}\left[{}^3D\ 3/2\right]^o_{1/2}$ and $4f^{13}5d6p\left({}^2F^o_{7/2}, {}^3F^o_2\right)_{5/2}$ states. The disagreements with the delayed-coincidence measurements (Blagoev *et al* 1978) are quite obvious as indicated in table 2. The origin might be connected to problems frequently encountered when using non-selective excitation. For all the present measurements, a nice agreement with the calculated values was found,

indicating that the HFR approach is suitable for correctly predicting the radiative properties of this singly ionized lanthanide.

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