Radiative lifetimes of excited W II levels

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Abstract. Radiative lifetimes of 19 selected W II levels with energies between 36 000 cm⁻¹ and 55 000 cm⁻¹ have been measured with the time-resolved laser-induced fluorescence technique. The ions are generated in a hollow cathode discharge and stored in a linear Paul trap. Selected states are populated with tunable dye laser pulses and the subsequent fluorescence is measured by means of a 5 Gigasample transient digitizer and a fast photodetector with a risetime of 700 ps. By taking into account both the temporal profile of the laser pulses and the separately measured response function of the system, the lifetime can be determined from the full decay curve. A refined evaluation procedure, taking into account saturation effects in the signals, reduces the uncertainty in our data to around 1%.

PACS. 32.50.+d Fluorescence; phosphorescence (including quenching) – 32.80.Pj Optical cooling of atoms; trapping

1 Introduction

We are on the way to measure reliable sets of atomic transition probabilities of tungsten lines for the first three spectra. The reason for that program is the renewed interest of the fusion community in tungsten as a divertor and limiter material [1,2]. In a first step we measured W I level lifetimes [3] and transition probabilities forming a set of 226 f-values [4] which is an enlargement of the data set in reference [5].

For W II we had to overcome the problem that the number of tungsten ions in the effusive beam extracted from our hollow cathode discharge (see Ref. [3]) was too small for a time-resolved laser-induced fluorescence (TLIF) measurement. We therefore developed a linear Paul trap for storing a number of ions sufficient for TLIF measurements [6]. We made a selection of a limited number of W II levels starting with the Kurucz atomic data base [7] and using the Moore tables [8] for assignment.

We have further improved our experimental equipment and the evaluation procedure. Now we are able to measure lifetimes with uncertainties of typically 1%. The lifetimes will be used for creating a set of W II oscillator strengths by combining them with the corresponding branching ratios we are going to measure.

2 Experimental arrangement

The linear Paul trap is part of our TRLIF experiment as described elsewhere [6]. The tungsten atoms and ions are generated in a modified version of our high-current hollow cathode [9,10]. The cylindrical cathode insert of 35 mm length and a central bore of 6 mm is stopped down to a 0.3 mm aperture on the low-pressure side. Within the discharge the buffer gas pressure is typically 300 Pa so that the trap is filled continuously through the aperture by pressure gradient. While the neutrals move through the trap as an effusive beam the tungsten ions are guided in an ion beam with a diameter of a few millimeters. For obtaining an efficient ion trapping the low-pressure side is filled with 3–40 Pa neon as a cooling gas. With the apparatus the fluorescence can be detected 4 cm behind the aperture or 15 cm behind. At the first distance also neutrals can be investigated. The discharge was operated in neon at currents between 0.3 A and 1 A.

A quanta ray DCR 11–3 Nd:YAG laser-pumped dye laser (Lambda Physik: LPD 3002) produces laser pulses of 4–5 ns duration (FWHM) with a spectral bandwidth of 0.2 cm−¹ and a repetition rate of 10 Hz. The spectral range 205–280 nm is obtained by frequency doubling with BBO and KDP crystals leading to a pulse duration of 3– 3.5 ns. The laser beam is crossed with the ion cloud/beam in the Paul trap. Perpendicular to the laser beam the fluorescence photons are imaged by a lens system onto the photocathode of a multiplier. The angle between the direction of polarization of the laser pulses and the direction of the detected fluorescence is set to the so-called magic angle of 54.7◦ to exclude the influence of collisional disalignment or hyperfine structure quantum beats [11] which can falsify the lifetime measurements. We need no longer optical filters for excluding unwanted light because scattered laser light is sufficiently suppressed by a stack of diaphragms and light from the discharge itself is excluded by the 0.3 mm aperture.

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Fig. 1. TLIF measurement on a W II level. The curve has been fitted considering saturation effects. Only part of the measuring points are shown in the figure.

Our multiplier (Hamamatsu R2496) has a finite risetime of 700 ps and a non-ideal response function with ringing due to the electrical circuitry. The ringing cannot be smoothed without a loss in time-resolution. We therefore measure the response function separately and include it in our evaluation procedure (see also Ref. [12,13]). In addition, we measure the temporal shape of our laser pulse by means of a fast photodiode (Hamamatsu R1328U–52) having a risetime of 60 ps. This arrangement is also used to trigger the fluorescence signal. Moreover, the shape of the laser pulse enters our evaluation procedure. The fluorescence curves are recorded time-resolved by means of a fast digitizing oscilloscope (Tektronix TDS 680 B) with an analog bandwidth of 1 GHz and a realtime scanning rate of 2×5 Gigasamples. This instrument allows the record of a full decay curve and simultaneously the record of the temporal laser pulse with a 200 ps time increment which is comparable with the risetime of the oscilloscope.

In the present experiment we typically added 500 single shots for obtaining a flawless S/N ratio, and we made at least 5 independent measurements on as many fluorescence transitions as possible.

3 Results and discussion

It is common practice to perform lifetime measurements at gas pressure as low as possible to exclude quenching collisions. We found, however, that pressures of the cooling gas between 3 Pa and 40 Pa are necessary for an efficient trapping of the ions. Below 3 Pa the cooling effect is too small, above 40 Pa the collisions begin to disturb trapping. We checked in a previous paper (see Ref. [6]) that in the pressure range used in our experiment pressure-dependent effects do not disturb our lifetime data.

Pumping a transition resonantly with a short laser pulse, i.e., with a huge number of photons, always leads to saturation effects in the fluorescence signals. The evaluation procedure as described in reference [12] has been

Table 1. Cu I radiative lifetimes.

Level	14	this work		
$4p^2P_{1/2}$	$7.27 + 0.06$	$7.36 + 0.09$		
$4p^2P_{3/2}$	$7.17 + 0.06$	$7.21 + 0.07$		

extended to handle saturation in the fluorescence signals by solving an appropriate rate equation model. The influence of the pulse shape of the laser and of saturation on the decay curves is demonstrated in Figure 1. The fitted curve matches the measuring points (only part of them are shown) in an excellent manner. When the saturation in our evaluation procedure is switched off the fit can no longer be done in a satisfactory way. We would like to state that if the laser pulses used are of comparable time duration to the lifetime to be measured saturation has to be included in order to avoid a systematic lengthening of the lifetime.

In order to test the quality of our TLIF apparatus and evaluation procedure we measured two prominent Cu I levels, $4p^2P_{1/2,3/2}$, and compared the lifetimes with the very accurate data from the Lund group [14]. As can be seen in Table 1 there is consistency within the mutual error bars, although the measuring procedures are quite different.

The W II lifetimes are given in Table 2 together with the standard deviation of the mean which is also around 1%. If the lifetime, however, is comparable with the pulse length of our laser the uncertainty can increase up to 3%. That is due to the shot-to-shot fluctuations in the laser pulses. We selected the 19 levels in such a way that the f-values of the most prominent W II lines in the spectral range 200–900 nm can be obtained from this lifetime data. A comparison with published values is difficult because W II data are scarce in the literature. A few of our levels have been measured by Kwiatkowski et al. [15]. The group is known for reliable measurements and the comparison confirms it. However, one level is out of the common range. We think the authors made a misidentification by measuring the lifetime from the line at 265.3425 nm which belongs to the upper level ${}^{4}P_{11/2}^{0}$ listed in Table 2 with a lifetime of 2.48 ns, instead of that from the 265.3564 nm line.

A more extended comparison can be obtained with the large Kurucz data base [7]. Unfortunately, there are two restrictions. The calculations are not of comparable accuracy as regards modern experimental data, and for complex atoms like tungsten the base gives only a limited number of lines collected from literature. Summing up the A values to find the lifetime for a particular level results in values that are systematically too large. Nevertheless, we applied the procedure and the result are given in Table 2 for comparison.

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Level	Energy	Transitions				Lifetimes (ns)		
	$\rm (cm^{-1})$		(nm) $\lambda_{exc.}$		$[7]$	[15]	this work	
${}^{6}F^{0}_{1/2}$	36165.35	276.4261				13.65 14.0 ± 0.7	14.42 ± 0.14	
	38576.32	259.1489	269.7706		9.09	11.3 ± 0.6	11.88 ± 0.07	
$\frac{{}^2S_{1/2}^0}{{}^6F_{3/2}^0}$	39129.41	255.4856	265.8032		10.02		13.94 ± 0.09	
$^6F^0_{5/2} \\ ^6F^0_{3/2}$	42049.45	246.6522	257.1449	267.7790	16.04		11.07 ± 0.19	
	42298.20	245.1475	255.5100		5.89		10.88 ± 0.13	
${}^{6}F_{7/2}^{0}$	42390.27	254.9101	265.3564		71.8	3.0 ± 0.3	76.1 ± 0.9	
$^6F^0_{5/2} \\ ^6F^0_{1/2}$	44354.82	242.7491	252.2041		16.37		9.90 ± 0.12	
	44455.18	224.8759	232.8312		58.8		2.67 ± 0.07	
${}^{6}F^{0}_{9/2}$	44758.10	249.6639	258.9165		28.34		21.63 ± 0.22	
	44877.18	239.7084	248.9235	258.1204	5.17		6.58 ± 0.15	
$^6F^0_{7/2} \\ ^6F^0_{3/2}$	44911.63	230.3818	266.6500		11.62		4.35 ± 0.06	
$^{6}F^{0}_{3/2}$	45553.70	227.0223			39.2		7.88 ± 0.24	
$\begin{array}{l} ^6F^0_{9/2}\\ ^6F^0_{3/2}\\ ^6F^0_{3/2} \end{array}$	46493.43	239.2926	247.7795		17.27		12.37 ± 0.10	
	47179.94	211.8875	259.8741		18.9		5.71 ± 0.08	
	47588.64	210.0675	248.8780		9.07		3.40 ± 0.11	
	48982.86	210.6189			20.6		5.00 ± 0.08	
$^6F^0_{3/2}\\ ^6F^0_{5/2}$	49242.10	209.4746	239.0376		24.1		3.33 ± 0.11	
${}^{6}F_{5/2}^{0}$	50292.33	212.1579			7.62		5.14 ± 0.06	
${}^4P^0_{11/2}$	54229.06	207.9122	265.3425		17.8		2.48 ± 0.08	

Table 2. W II radiative lifetimes

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