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Measurements of excited state lifetimes and oscillator strengths in Pt II

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Abstract. We report on radiative lifetime measurements of short-lived states in singly ionized platinum by excitation with pico-second laser pulses and subsequent time-resolved detection. The upper levels were populated from metastable ion levels in a laser-generated plasma using radiation in the UV/VUV spectral range. The following lifetimes were obtained: $\tau(5d^8(^3F_4)6p_{1/2}, J = 7/2) = 3.9(3)$ ns, $\tau(5d^8(^3F_4)6p_{3/2}, J = 9/2) = 3.5(3)$ ns and $\tau(5d^8(^3F_3)6p_{1/2}, J = 5/2) = 2.6(3)$ ns. By using relative intensities of the lines found in the literature, oscillator strengths were determined.

1. Introduction

Following the launch of the Hubble Space Telescope in 1990 there has been a large number of observations of stellar spectral lines in the UV/VUV spectral range. These belong to a large number of different atoms and ions. Of special interest has been the investigation of chemically peculiar stars such as χ Lupi (Leckrone *et al* 1993). In these stars there is an overabundance of heavy elements such as mercury. In order to understand the formation of such stars, it is interesting to investigate the abundance of many other heavy elements such as ruthenium, gold, palladium and platinum. All these elements have been observed in high abundance in this star. To deduce the abundance, oscillator strengths must be known. One way of achieving this is to combine measurements of branching fractions with measured excited-state lifetimes. This was carried out in a recent study on ruthenium (Johansson *et al* 1994). In that paper, a general-purpose technique for the production of free ions in a laser-produced plasma was presented. In order to be able to measure the lifetimes of the relevant high-lying states in platinum we have investigated and exploited the possibility of performing excitation from high-lying metastable states. Excitation from levels as high in energy as $13\,329$ cm^{-1} was used to measure high-lying states ($51\,408$–$64\,757$ cm^{-1}). To enable us to make accurate measurements of the short lifetimes we have employed laser pulses of short duration (<100 ps), and detection of the fluorescence radiation with a high temporal resolution (<1 ns). The exciting source had a sufficiently small bandwidth (<1 cm^{-1}) to allow for selective excitation. Our technique enabled us to make measurements with an accuracy of about 10%.

2. Experimental set-up

The experimental set-up is shown in figure 1. In order to obtain laser pulses of relatively short duration but with a narrow spectral bandwidth, a distributed-feedback dye laser

(DFDL) oscillator (Schank *et al* 1971, Bor and Schaefer 1983) was pumped by a small fraction of the output from a mode-locked and Q-switched Nd:YAG laser. Two dye amplifiers pumped by the same frequency-doubled Nd:YAG laser were used in order to obtain tuneable radiation around 750 nm with a pulse energy just above 1 mJ. The radiation from the dye laser was subsequently mixed with the fourth harmonic from the Nd:YAG laser in a β-barium borate (BBO) crystal, yielding the desired output at 194 nm. The pulses at this wavelength had an energy of a few tens of μJ. The DFDL oscillator provides an efficient way of obtaining narrow-band, wavelength-stable operation of a pico-second laser (Schade *et al* 1990).
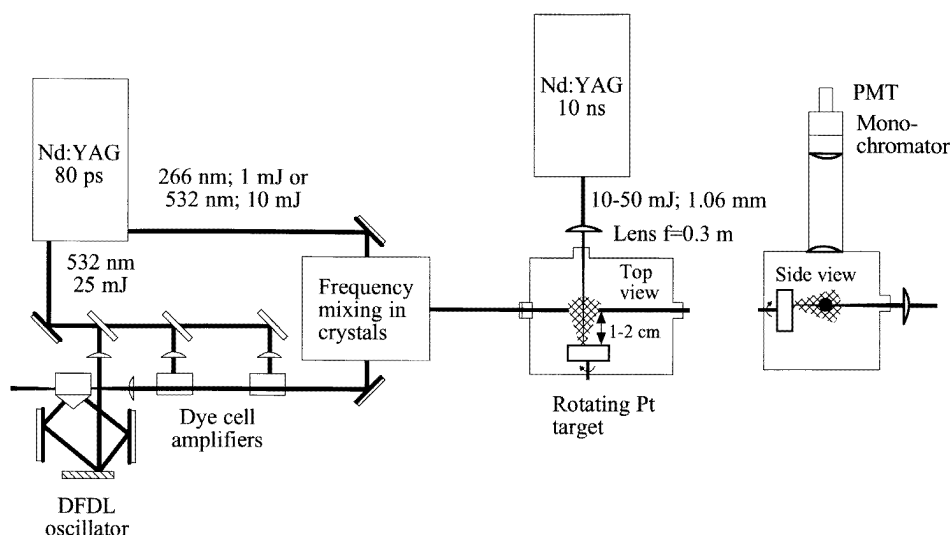

Figure 1. Experimental set-up for measuring short lifetimes in a laser-produced plasma.

Free platinum ions were obtained in a laser-produced plasma. The output from a Q-switched Nd:YAG laser producing pulses of 10 ns duration and an output energy of 10–50 mJ was focused with a 30 cm focal-length lens onto a rotating platinum target. Effective population of metastable states persisted until the plasma density was sufficiently low to allow for perturbation-free measurements.

The high-lying ionic states to be studied were excited with the tuneable short-pulse radiation which was directed perpendicularly to the direction of flight of the ions. The fluorescence was detected in a direction perpendicular to the direction of flight as well as to that of the exciting radiation. Apart from the rapidly decaying laser-induced fluorescence, a background of plasma emission with a duration of several μs was observed. To single out the desired radiation and to reduce this continuous background radiation from the plasma, a 0.25 m spectrometer with about 4 nm/mm dispersion was used.

The detector was a Hamamatsu 1564U microchannel-plate PMT which was connected to a Tektronix DSA 602 digital oscilloscope with an analogue bandwidth of 1 GHz and a sampling rate of 2 Gsample/s. Data were also acquired with interleaved sampling with 5 Gsamples/s. It was found, however, that data acquisition was more efficient with real-time sampling. This is due to pulse-to-pulse fluctuations from the laser source which influence interleaved sampling measurements. The oscilloscope was optically triggered with a jitter below 100 ps. The total response of the system was mainly limited by the input amplifier

of the oscilloscope. Some influence from the limited rise-time of the PMT (200 ps) and the signal cables is also expected. The measured FWHM of the stray light from the short-pulse laser as observed with this detection system was below 1 ns.

Table 1. log(gf)-values for the transitions from the three states for which lifetimes were measured. The lifetimes from this study and the line intensities from Sansonetti *et al* (1992) were used. The states are denoted according to Moore (1971).

Lower level energy (cm^{-1})	Transition wavenumber (cm^{-1})	Intensity	A (s^{-1})	log(gf)
$5d^8(^3F_4)6p_{1/2}$, $J = 7/2$; 51 408.370 cm^{-1}; $\tau = 3.9(3)$ ns				
4786	46 622	350 000	2.31 E+08	0.10
9356	42 052	23 000	1.52 E+07	−0.99
13 329	38 079	5100	3.36 E+06	−1.56
16 820	34 587	2600	1.71 E+06	−1.77
18 097	33 311	6800	4.48 E+06	−1.31
$5d^8(^3F_4)6p_{3/2}$, $J = 9/2$; 60 907.688 cm^{-1}; $\tau = 3.5(3)$ ns				
4786	56 121	93 000	1.62 E+08	−0.11
9356	51 551	53 000	9.21 E+07	−0.28
18 097	42 810	8800	1.53 E+07	−0.90
24 879	36 028	7900	1.37 E+07	−0.80
29 261	31 646	1800	3.13 E+06	−1.33
$5d^8(^3F_3)6p_{1/2}$, $J = 5/2$; 64 757.343 cm^{-1}; $\tau = 2.6(3)$ ns				
8419	56 338	86 000	1.32 E+08	−0.43
9356	55 401	3200	4.93 E+06	−1.84
13 329	51 428	63 000	9.70 E+07	−0.48
15 791	48 966	62 000	9.55 E+07	−0.45
16 820	47 936	6900	1.06 E+07	−1.38
18 097	46 660	3900	6.01 E+06	−1.61
21 168	43 589	1800	2.77 E+06	−1.88
23 461	41 296	18 000	2.77 E+07	−0.84
23 875	40 882	1100	1.69 E+06	−2.04
29 030	35 727	1000	1.54 E+06	−1.96
32 237	32 520	380	5.85 E+05	−2.30
36 484	28 273	2500	3.85 E+06	−1.36

3. Measurements and results

Lifetimes for three states in Pt II were measured. They were all in the range 2–4 ns. The states were chosen because strong lines observed in stellar spectra originate from them. The results can be found in table 1 and a decay curve for one of the states is given in figure 2. Since the spectral resolution of the detection system was as low as 2 nm, a non-negligible plasma background was present. This problem was avoided by acquiring data with and without the dye laser radiation and subsequent subtraction of the time-resolved curves. In principle, the radiative lifetime could now be deduced by fitting an exponential to the recorded fluorescence. In order to avoid any influence from the detection system on the lifetime data another approach was chosen. The lifetimes were evaluated by fitting a convolution of an exponential and a laser pulse as registered by the detection system to

the recorded fluorescence. This made it possible to evaluate the lifetimes with an accuracy as good as 8% for the longest lifetime. In order to investigate systematic effects from multiple scattering or collisions, the fluorescence was recorded for different delays between the plasma-generating laser and the excitation pulse. For the $64\,757$ cm^{-1} level, which was excited from the highest-lying metastable state, the delay was varied between 0.8 and 1.8 μs. No systematic influence on the lifetimes could be seen. For excitation from the lowest metastable level a delay of up to 5 μs gave a sufficiently strong signal to allow for a reliable lifetime determination. The distance between the platinum target and the interaction region in the plasma plume, usually 5 mm, was also independently varied without any influence on the deduced lifetimes. Since the studied states have short lifetimes, no problems due to flight-out-of-view effects occur although the expansion velocities are of the order of 10 km s^{-1}.
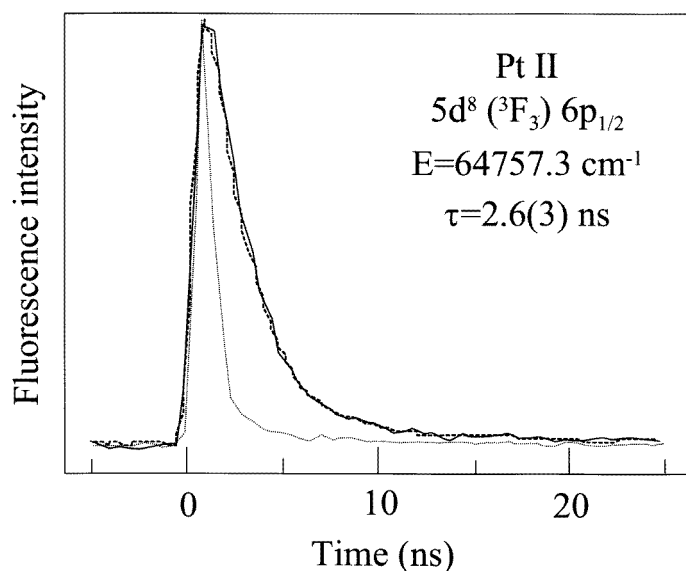

Figure 2. Experimental recording of the fluorescence from a high-lying state in ionized platinum (solid). The laser pulse (dashed) and a convolution of the laser pulse and an exponential with a decay time of 2.6 ns (dotted) are also shown.

By using the relative intensities of a large number of spectral lines in the UV region measured by Sansonetti *et al* (1992) the log(gf)-values could be deduced. These values are given in table 1. Transitions in the IR/visible spectral region to states in the $5d^7 6s^2$ configuration are allowed. In our experiments we inspected the visible spectral range without observing any of these transitions. This is in agreement with *ab initio* calculations which also yield that the intensity of IR transitions are negligible.

No error bars were given for the relative intensities. It should be noted, however, that for the transition at $46\,622$ cm^{-1} the branching fraction is almost 90%. This means that even a large error in the relative intensity will give a small deviation in gf-value. As an example, an error of factor three in relative intensity gives a 15% deviation in gf-value. For the other two measured lines, the branching fractions are more evenly distributed and a certain error (given in per cent) in the intensity will give roughly the same error in the gf-value.

4. Discussion

In this paper we have measured radiative lifetimes for upper states of transitions that have been observed in stellar spectra. By combining these data with branching ratios, log(gf)-values have been obtained.

We have shown that perturbation-free measurements of radiative lifetimes of states lying as high as $64\,757$ cm^{-1} above the ground state can be performed with selective one-step laser excitation. Lifetimes as short as 1 ns can be measured with the present set-up. The laser system can be operated between 400 and 900 nm. Frequency mixing in crystals can readily be performed. The ion sources described in this and in a previous study (Johansson *et al* 1994) are suitable for the production of any atom or ion which is available as a bulk, powder or salt.

Presently, the excitation wavelengths are limited by the absorption cut-off of BBO at 190 nm. In order to extend this to even shorter wavelengths, frequency-mixing in gases can be used (Borsutzky *et al* 1990). For wavelengths longer than 110 nm phase matching can be obtained and it is expected that the photon yield will be high enough to allow for fluorescence detection. For even shorter wavelengths where the photon yield is even smaller it might be possible to apply the photoionization pump-probe technique which was recently demonstrated on the resonance line of neutral helium (Larsson *et al* 1995).

References

Bor Z and Schaefer F P 1983 *Appl. Phys.* B **31** 209
Borsutzky A, Brünger R and Wallenstein R 1990 *Applied Laser Spectroscopy* ed W Demtröder and M Inguscio (New York: Plenum) p 63
Johansson Se, Juoeizadeh A, Litzén U, Larsson J, Persson A, Wahlström C-G, Svanberg S, Leckrone D S and Wahlgren G M 1994 *Astrophys. J.* **421** 809
Larsson J, Mevel E, Zerne R, L'Huillier A, Wahlström C-G and Svanberg S 1995 *J. Phys. B: At. Mol. Opt. Phys.* **28** L53
Leckrone D S, Johansson Se, Wahlgren G M and Adelman S J 1993 *Phys. Scr.* T **47** 149
Moore C 1971 *Atomic Energy Levels* NSRDS NBS **35** vol III (Washington, DC: US Government Printing Office)
Sansonetti J, Reader J, Sansonetti C and Acquista N 1991 *J. Res. Natl. Inst. Stand. Technol.* **97** 1
Schade W, Garbe B and Helbig V 1990 *Appl. Opt.* **29** 3950
Schank C V, Bjorkholm J E and Kogelnik H 1971 *Appl. Phys. Lett.* **18** 395