The FERRUM project: radiative lifetimes of the 3d(5)(S-6)4s4p(P-3)y P-6 degrees states of FeII measured with time-resolved vacuum ultra-violet laser spectroscopy

Li, Z. S; Lundberg, Hans; Berzinsh, U; Johansson, S; Svanberg, Sune

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The FERRUM project: radiative lifetimes of the $3d^5(^6S)4s4p(^3P)y\ ^6P^\circ$ states of Fe II measured with time-resolved vacuum ultra-violet laser spectroscopy

Z S Li†, H Lundberg†, U Berzinsh,†§, S Johansson‡ and S Svanberg†

† Department of Physics, Lund Institute of Technology, PO Box 118, S-221 00 Lund, Sweden
‡ Atomic Spectroscopy, Department of Physics, Lund University, PO Box 118, S-221 00 Lund, Sweden

E-mail: Zhongshan.Li@fysik.lth.se

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Abstract. We report on lifetime measurements of the $3d^5(^6S)4s4p(^3P)y\ ^6P^\circ$ states of Fe II using time-resolved vacuum ultraviolet (VUV) laser spectroscopy. A laser-produced plasma has been used as the source of free Fe^+ ions. The tunable VUV radiation was obtained employing resonantly enhanced sum-difference four-wave-mixing of short laser pulses. We obtained $\tau(^6P^\circ_{3/2}) = 3.90(20)$ ns, $\tau(^6P^\circ_{5/2}) = 3.80(20)$ ns and $\tau(^6P^\circ_{7/2}) = 3.65(20)$ ns.

(Some figures in this article are in colour only in the electronic version; see www.iop.org)

1. Introduction

Due to the high cosmic abundance and the richness in spectral lines, Fe II forms the most common spectrum in astrophysics. The quantitative analysis of astrophysical spectra of stars, free interstellar medium, nebulae etc relies very much on the quality of the atomic data. The aim of the FERRUM project is to extend and improve the database of the oscillator strengths of Fe II by combining radiative lifetimes and experimental as well as theoretical branching fractions. The lifetimes are measured in the Lund Laser Centre (see Svanberg *et al* (1994)) with a time-resolved laser spectroscopic method and the experimental branching ratios have been obtained with the Fourier transform spectrometer (FTS) for $180 < \lambda < 800$ nm at Lund University.

Many efforts have been made in extending the Fe II transition probability database. The previous work has been summarized by Li *et al* (1999a) in the first FERRUM project publication. In that paper, the cascade-free laser spectroscopic lifetime measurements of two intermediate-excited states $z\ ^4S^\circ_{3/2}$ and $y\ ^4P^\circ_{5/2}$ of the $3d^64p$ configuration were reported for the first time. In a second paper, Sikström *et al* (1999) published the absolute oscillator strength of 18 Fe II lines from highly excited $3d^64p$ levels, which were obtained by combining the experimental lifetime values with the measured branching ratios. In a third paper, six levels of the $3d^64d$ configuration were measured with two-step excitation from the ground state. By combining the lifetimes with the FTS branching ratio results, Nilsson *et al* (2000) reported oscillator strengths of 29 4p–4d lines.

§ Permanent address: Institute of Atomic Physics and Spectroscopy, University of Latvia, Latvia.

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In the previous FERRUM project papers, effort was put into obtaining reliable f-values of weak transitions or transitions from high-lying states in order to circumvent the line saturation problem in stellar spectra due to the high cosmic elemental abundance of iron. Also, by using Fe II lines from different excitation energies in abundance studies, one can check the validity of assuming thermal equilibrium in stellar model atmospheres. In this paper, we report the lifetime measurements of the levels of the $3d^5(^6S)4s4p(^3P)y\,^6P^o$ term of Fe II. These levels couple strongly to the fine-structure levels of the ground term. These ground-term transitions of Fe II are the primary lines in the neutral hydrogen region of the interstellar medium, and are suitable for obtaining an accurate interstellar iron column density (see Savage *et al* (1993)). In this paper, time-resolved laser-induced fluorescence was used for the lifetime measurements. A laser-produced plasma was employed as the source of free Fe^+ ions. The required vacuum ultraviolet (VUV) excitation was accomplished by using resonantly enhanced sum-difference four-wave mixing of two laser beams in krypton gas. In order to be able to measure the short lifetimes, one of the lasers was pumped with pulses from a Nd:YAG laser, which were temporally compressed using stimulated Brillouin scattering (SBS). The resultant VUV laser pulses had a duration of about 1 ns. Lifetime values were evaluated from the time-resolved fluorescence signal recorded by a fast detection system.

2. Experimental setup

A partial energy level diagram of Fe II, relevant to this experiment, is shown in figure 1. The $y\,^6P^\circ$ levels were excited from the $a\,^6D$ ground-term fine-structure levels. The required VUV excitation laser pulses at about 161 nm were produced through a two-photon resonantly enhanced sum-difference four-wave-mixing process as shown in the insert in figure 1. Through collinear phase matching in a Kr gas, two UV beams ω_R and ω_T stimulated coherent VUV radiation at $\omega_{VUV} = 2\omega_R - \omega_T$. The conversion efficiency was substantially enhanced by tuning the λ_R to 212.55 nm, which is in two-photon resonance with a $4p^6$–$4p^55p$ transition.

The setup of the VUV laser system is shown in figure 2. The green pumping laser power comes from two frequency-doubled injection-seeded Nd:YAG lasers (Continuum NY-82) with a pulse duration of about 8 ns. Two dye lasers (Continuum ND-60), which operated on DCM dye, were pumped separately. Before entering the dye laser, one green beam was compressed in a SBS unit (for technical details see Li *et al* (1999b)) to about 1 ns. The output from the dye laser, which was pumped by the short laser pulse, was fixed to 637.65 nm. This beam was frequency tripled in an optical crystal system, which includes a BBO crystal, a KDP crystal and a retarding plate (for technical details see Bengtsson *et al* (1990)), and yielded $\lambda_R = 212.55$ nm laser radiation of about 1 ns duration. The output from the other dye laser was frequency doubled in a KDP crystal, and the second harmonic, which is shown in figure 1 as λ_T, can be tuned freely. The two UV laser beams were combined with a dichroic mirror and focused together in the krypton cell with an achromatic lens. The two Nd:YAG lasers were externally triggered from the same delay unit to achieve not only the spatial but also the temporal overlap. The VUV laser emission at λ_{VUV} was tunable by varying λ_T. The VUV laser beam was separated from the pump beams with a LiF prism and sent through vacuum pipes to the vacuum chamber, where it crossed the Fe^+ ionic beam.

The experimental setup is schematically illustrated in figure 3. A small Nd:YAG laser (Continuum Surelite) provided a green beam of 10 ns pulse duration and about 5 mJ pulse energy. This laser was focused through a glass window onto a rotating iron target in the vacuum chamber, where a small plasma explosion was introduced in the focus point of the iron target surface after each laser pulse. Free Fe^+ ions were observed in the cooling plasma a little while after the explosion. All the Nd:YAG lasers were externally triggered, which

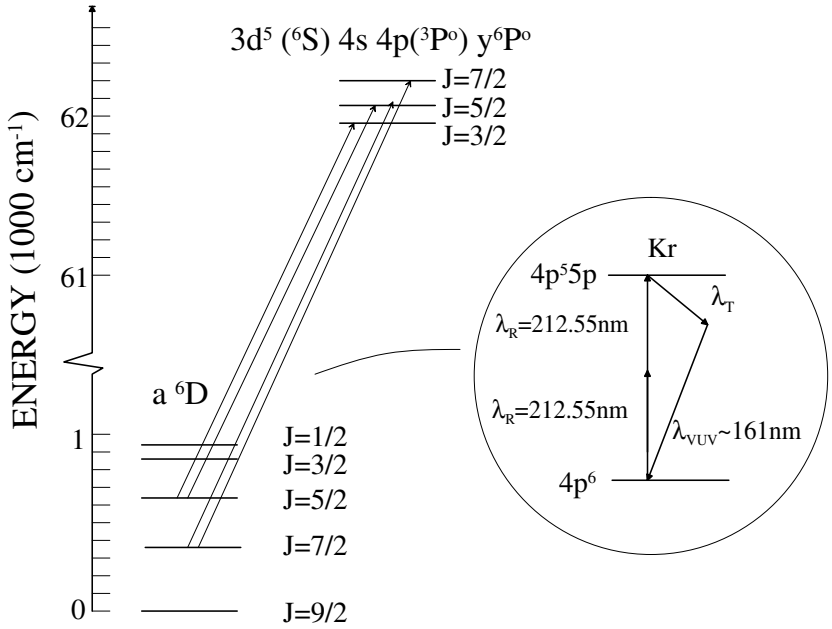

Figure 1. Partial energy level diagram for Fe II relevant to this experiment, and the generation scheme for the VUV radiation shown as an insert.

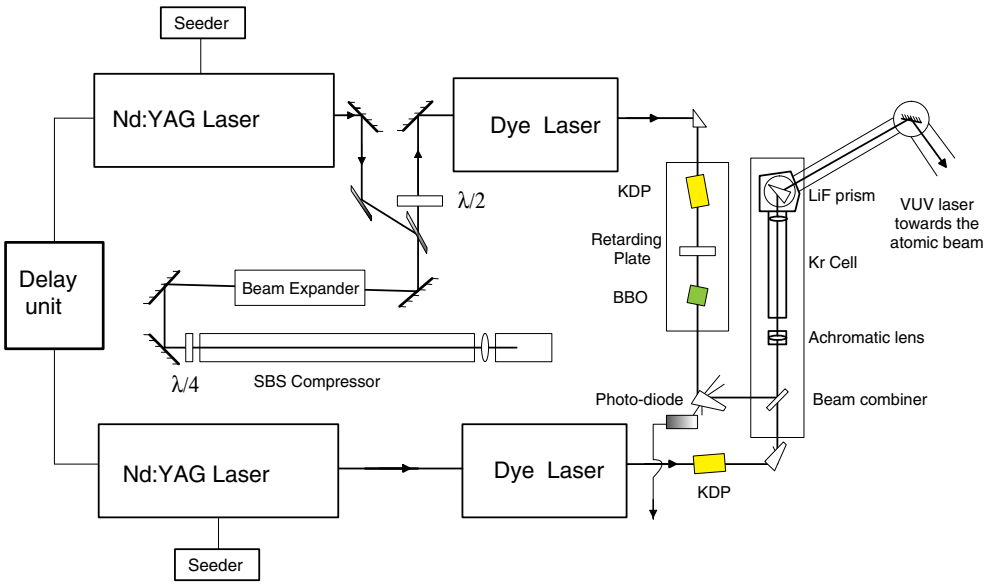

Figure 2. Schematic illustration of the VUV laser system.

enabled full control of the delay time between the ablation and excitation pulses. The VUV laser beam was sent through the plasma about 10 mm above the target surface. By tuning the wavelength of the VUV excitation laser, the proper upper levels of the illuminated Fe^+ ions in the plasma were selectively excited. A MgF lens was used to image the interaction region to

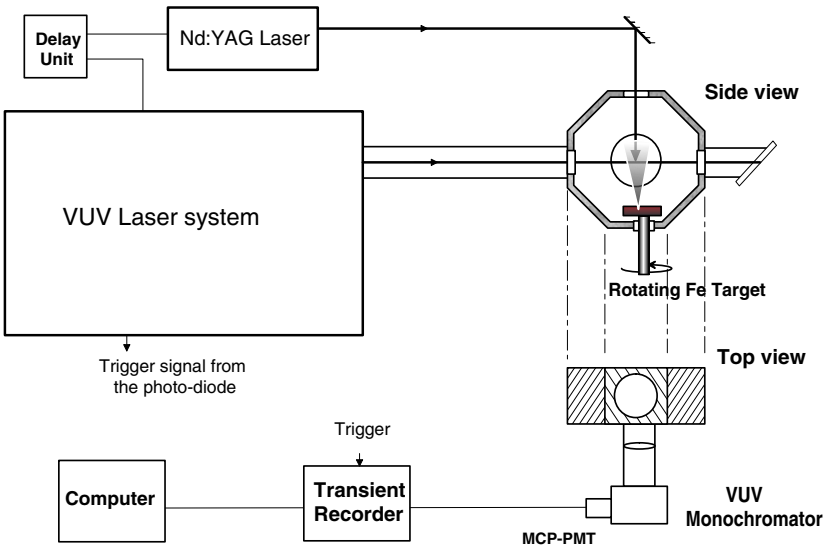

Figure 3. Schematic illustration of the experimental setup used in this paper.

the input slit of a VUV monochromator (Acton Model VM502). The spontaneous emission line of the same transition as the excitation was chosen to record the exponential decay. The fluorescence photons emitted from the excited Fe^+ ions were selected by the monochromator and detected by a Hamamatsu R3809U-58 microchannel-plate photomultiplier tube (rise time 170 ps). The transient signal output from the detector was recorded by a Tektronix TDS 684B transient oscilloscope (bandwidth 1 GHz and real-time sampling rate 5 Gsample s^{-1}), which was triggered by a Thorlabs SV2-FC photo-diode (rise time 120 ps) driven by the short compressed laser pulse. The recorded fluorescence signals were transferred to an IBM personal computer, where the evaluation of the lifetime values could be performed in direct connection with the measurements.

3. Measurements and results

A laser-produced plasma has proved to be a versatile source of target atoms in time-resolved laser-induced fluorescence experiments. In the plasma, not only free neutral atoms (see Li *et al* (1998)), but also sufficiently high densities of singly and doubly ionized ions (Li *et al* 1999c) in the ground or metastable states can be produced, which makes the excitation quite flexible. By simply changing the delay time between the atomization pulse and the excitation pulse, the focus size, the atomization laser pulse energy and the height of the interaction point above the target surface, such plasma parameters as temperature and density in the excitation zone can be varied. The relative density of neutral atoms and ions of different ionization stages and metastable states can be chosen by varying the above parameters. Proper plasma conditions have been carefully chosen for the final measurements.

In a previous FERRUM project paper (Li *et al* 1999a), excitation from the high-lying metastable state $a\,^4P_{5/2}$ (13 474 cm^{-1}) was utilized in order to excite the $y\,^4P^\circ_{5/2}$ and $z\,^4S^\circ_{3/2}$ states, in which high-density and -temperature plasma was required. In the present experiment, the excitations are from the fine-structure levels $a\,^6D_{5/2}$ and $a\,^6D_{7/2}$, which means that relatively cool and less dense plasma can be used, and that collisional effects are reduced. However,

Table 1. Radiative lifetime measurements of C I.

Level	Energy (cm^{-1})	Lifetime (ns)	Previous results
$2s^2 2p3s\ ^3P_0$	60 333.43	3.00(20)	
$2s^2 2p3s\ ^3P_1$	60 352.63	3.05(20)	3.1(2)[a]
$2s^2 2p3s\ ^3P_2$	60 393.14	3.05(20)	2.9[b]

[a] Haar *et al* 1991 (experimental results).
[b] Hibbert *et al* 1993 (calculations).

Table 2. Experimental radiative lifetimes of Fe II.

			Excitation	
Level	Energy (cm^{-1})	Lifetime (ns)	Lower level (cm^{-1})	Wavelength (nm)
$3d^5(^6S)4s4p(^3P^\circ)y\ ^6P_{3/2}$	61 974.933	3.90(20)	667.683	163.11
$3d^5(^6S)4s4p(^3P^\circ)y\ ^6P_{5/2}$	62 049.025	3.80(20)	384.790	162.17
			667.683	162.92
$3d^5(^6S)4s4p(^3P^\circ)y\ ^6P_{7/2}$	62 171.615	3.65(20)	384.790	161.85

since the VUV laser beam is relatively weak, a high density of Fe^+ was needed in order to get a reasonably high signal in the measurements. Thus, radiative trapping effects, which could increase the measured lifetime values, have to be considered. A careful check of this effect by varying the delay time between the atomization and excitation laser pulses was performed. As the delay varied from 1 to 4 μs, the observed fluorescence signal changed from 30 to 3 mV, while no systematic effects of the evaluated lifetime values were observed. This justified the fact that the radiation trapping effects were neglected under the present experimental condition.

The polarization direction of the λ_{VUV} beam is the same as that of the λ_T beam. By changing the polarization direction of the λ_T with a double Fresnel rhomb, the λ_{VUV} polarization could be changed freely. In the experiments, the polarization direction of the λ_{VUV} beam was set to 54.7°, the so-called magic angle, with regard to the detection direction. This relaxes not only the hyperfine-structure quantum beat problem, but also the elastic collisional disalignment effects on the lifetime measurements (Hannaford *et al* 1983).

To test the whole experimental system in a comprehensive way, the well known lifetimes of the C I $2s^2 2p3s\ ^3P$ fine-structure levels were measured with the present setup. The results were compared with the most recent experimental (Haar *et al* 1991) and theoretical (Hibbert *et al* 1993) results, see table 1. Good agreements were achieved.

During the lifetime measurements of Fe^+, similar conditions as those for C I studies were adopted. The temporal shape of the excitation laser pulse was recorded by detecting the direct scattered light of the excitation beam from a metal rod inserted into the interaction region. Lifetime values were evaluated by fitting the recorded fluorescence signal to a convolution of the recorded laser pulse with an exponential decay. In order to keep the detection system in the linear response region, only very weak signals were recorded in the measurements. The recorded curves were averaged for 1000–4000 pulses to get an adequate signal-to-noise ratio. A typical fluorescence signal from the $y\ ^6P^\circ_{5/2}$ level, the excitation laser pulse shape and a convolution fitting are shown in figure 4. More than 20 pairs of pulses and signals of each level were recorded and evaluated. The mean values are shown as the final results in table 2, where the quoted error bars represent not only the random scattering of different measurements, but also a conservative estimation of possible systematic errors.

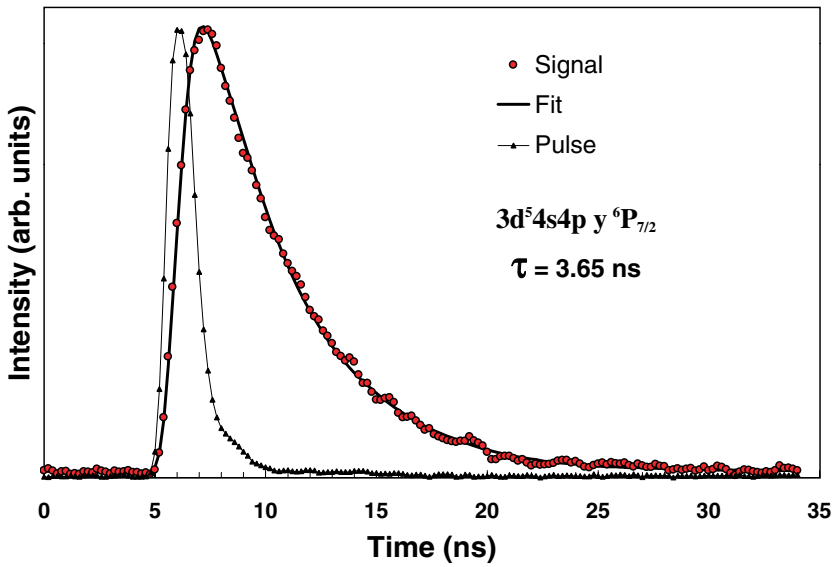

Figure 4. A typical recording of the fluorescence signal, the laser pulse and a fitting to the signal with a convolution between the laser pulse and a pure exponential.

4. Discussion

As mentioned in the introduction, the Fe II multiplet UV 8, $a\,^6$D–$y\,^6$P°, around 160 nm is of great importance for astrophysical applications, and accurate atomic data are therefore useful. For cool interstellar gas this is the only prominent Fe II multiplet in the wavelength region below 200 nm accessible to the Hubble space telescope. It is therefore natural that the target of the first direct measurements of oscillator strengths (f-values) for Fe II lines below 200 nm was the $a\,^6$D–$y\,^6$P° multiplet, performed by Mullman *et al* (1997) using the synchrotron radiation facility at Wisconsin–Madison. These authors report absolute f-values for seven of the nine possible transitions. In a later paper Donnelly and Hibbert (1999) present theoretical data for the same lines and make a comparison between experiment and theory. In table 4 of that paper comparisons are also made with the frequently used calculations of Kurucz (1988) and with the recent calculations of Raassen and Uylings (1998).

In order to compare our data with previous data we need to convert the f-values to transition probabilities. We also need to include all strong transitions from a given upper level to derive the radiative lifetime by inverting the sum of the transition probabilities. The experimental data from Mullman *et al* (1997) contain f-values for all three transitions from the $J = 3/2$ level of $y\,^6$P°, but only for two transitions from the $J = 5/2$ and $7/2$ levels. The same is true for the theoretical data from Donnelly and Hibbert (1999), but f-values for the 'missing' lines of UV 8 from the $J = 5/2$ and $7/2$ levels are available in Donnelly (1999). There are still differences between the theoretical data listed in table 3. The lifetimes derived from the calculations of Raassen and Kurucz include transitions to all possible lower levels, whereas the lifetimes of Donnelly and Hibbert only include the UV 8 lines. This introduces a correction that decreases these lifetimes by about 5%. From these considerations, it turns out that the calculations by Donnelly and Hibbert agree extremely well with our measurements. The calculations by Raassen and Uylings (1998) and by Kurucz (1988) agree within 10–15% with the measurements, which is also very good for complex spectra.

Table 3. Comparison of lifetimes derived from theoretical and experimental f-values.

		Lifetime (ns)				
Level (cm^{-1})	J	This work	D and H	R and U	K	M
61 974.933	3/2	3.90(20)	4.14	4.49	3.55	4.8
62 049.025	5/2	3.80(20)	3.95	4.36	3.35	(4.78)
62 171.615	7/2	3.65(20)	3.77	4.10	3.18	(4.12)

D and H: Donnelly's PhD Thesis (1999).
R and U: Raassen and Uylings (1998).
K: Kurucz (1988).
M: Mullman *et al* (1997), see text.

The lifetime derived from the experimental f-values of Mullman *et al* (1997) of the three UV 8 lines from the $J = 3/2$ level is 23% longer than our value. It is, of course, expected to be longer since other transitions from the upper level not belonging to the UV 8 multiplet are not included. However, the value is also 16% higher than the value derived from the data of Donnelly and Hibbert (1999), which is based on the same three lines as the Mullman *et al* measurements. As pointed out by Donnelly and Hibbert (1999) and shown in table 4 of their paper, there is a good agreement between measured and calculated f-values for four of the seven lines but a slight disagreement for the remaining three lines. We have briefly looked at our uncalibrated emission FTS spectra recorded at Imperial College (London) in the 160 nm region (used for wavelength measurements), and we also find a deviation from the previous experimental values for the same three lines, but a general good correlation with the calculations.

There is a good agreement in the ratios of the three sets of calculated oscillator strengths of UV 8, as shown in table 5 in Donnelly and Hibbert (1999), but deviations in the absolute values. These deviations result in the difference in the lifetimes in table 3. A reasonable explanation of these deviations is how the mixing of the $y\,^6P^\circ$ level is accounted for in the calculations. A small difference in the level composition may easily cause a substantial difference in the f-values. It is known from laboratory analyses (Johansson 1978) that the $y\,^6P^\circ$ levels are mixed with close-lying 4p levels of the same J-value but of a different configuration. Considering the nature of this type of level mixing in complex spectra, which is due to accidental coincidences in energy between levels of the same J-value, it is remarkable that the calculations agree so well with observations. There is experimental evidence that the $y\,^6P^\circ$ levels are mixed with both doublets and quartets.

5. Conclusions

We have reported experimental lifetimes for the astrophysical important $y\,^6P$ term of Fe II, which gives rise to the frequently observed UV 8 multiplet around 160 nm in satellite spectra of various astrophysical sources. To our knowledge, this is the first time that the complicated combination of an ionic species, a short lifetime and VUV laser excitation has been achieved. The lifetimes have been compared with three different theoretical calculations and one measurement of the oscillator strengths of the UV 8 multiplet. In order to get a proper comparison of transition rates, the lifetime experiment should be followed by measurements of the branching fractions. As the strongest lines appear in the 160 and 258 nm region this must be done with a VUV FTS.

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