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The FERRUM project: radiative lifetimes of intermediate-excitation states of Fe II measured in a fluorescence signal induced by laser pumping from a metastable state

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Abstract. A new collaborative program, the FERRUM project, aims at extending and improving the database of oscillator strengths for the astrophysical important Fe II spectrum. In this paper we report on the experimental method applied for measurements of radiative lifetimes of Fe II levels at intermediate excitation (5-10 eV) that can be populated by laser excitation from metastable states. The technique involves a laser-produced plasma as a source for metastable ions, a stimulated Brillouin scattering cell for shortening the laser pulse, and fast time resolved detection. We have applied the technique on two Fe II levels at about 7.5 eV and obtained the lifetimes: $\tau(z^4S^0_{3/2}) = 3.8(2)$ ns; $\tau(y^4P^0_{5/2}) = 3.6(3)$ ns.

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1 Introduction

Absorption spectra of stars at temperatures of about 10000 K contain numerous Fe II lines from the far ultraviolet to the near-infrared wavelength regions. This is due to the high cosmic abundance of iron, the ionization balance in stellar atmospheres, and the richness in spectral lines of the complex atomic system. The range of excitation energy of Fe II levels that have been observed in absorption spectra extends up to 12-13 eV. Due to the high density of energy levels and lines, Fe II provides possibilities to test the plasma conditions in the line forming regions. These conditions are critical for the validity of stellar model atmospheres, used for the determination of the chemical composition of the star's outer layers. Most models assume local thermal equilibrium (LTE), and the abundance is derived by the application of the Boltzmann distribution and the Saha equation.

Determination of an elemental abundance requires unblended stellar lines, for which accurate oscillator strengths (f-values) are available. The f-values used for Fe II are to a great extent calculated from theoretical codes, and their uncertainty can hardly be estimated. Until now, the experimental f-values are limited to transitions, where the upper level belongs to the $3d^6(^5D)4p$ subconfiguration, and the lower level has an excitation potential of less than 4 eV. These f-values have been obtained by combining experimental radiative lifetimes with measured branching fractions. The latter measurements are performed in emission and are complicated by an uncertain degree of self-absorption. Also, strong lines from low-excitation levels are often saturated in the stellar spectrum, and are less appropriate for abundance studies. Weaker lines either from levels at higher excitation energy or with a small branching fraction are preferred. Our FERRUM project aims at obtaining experimental data for such transitions.

The atomic structure of the Fe^+ ion has been extensively studied by Johansson [1]. Some branching fraction measurements have been performed by Pauls *et al.* [2] and Heise *et al.* [3]. Quite some effort has been put into measurements of Fe II lifetimes in the past 25 years. Based on non-selective excitation processes, beam foil measurements were performed by Smith *et al.* [4], Dolby *et al.* [5] and Johansson *et al.* [6]; electron beam excitation measurements were performed by Assousa *et al.* [7] and Brzozowski *et al.* [8]. The first laser spectroscopic lifetime measurement was published by Hannaford *et al.* [9] in 1983. Radiative lifetimes of the LS terms $z\ ^6D^0$, $z\ ^4F^0$, $z\ ^4D^0$ and $z\ ^4P^0$ were determined using laser-induced fluorescence in a sputtered ion vapor. With the improvement of laser techniques, the same group repeated their old measurements with the same experimental scheme and measured lifetimes of $z\ ^6F^0$ and $z\ ^6P^0$ in 1992 [10]. Laser excitation with a delayed coincidence detection technique was used by Schade *et al.* [11] to determine the lifetimes of the terms $z\ ^6D^0$, $z\ ^6F^0$ and $z\ ^6P^0$ in 1988. Biémont *et al.* [12] remeasured the lifetimes of the same terms with the beam-laser technique in 1991, and these measurements were extended to levels of the terms $z\ ^4D^0$, $z\ ^4F^0$ and $z\ ^4P^0$ by Guo *et al.* [13] in 1992. In the present paper we report for

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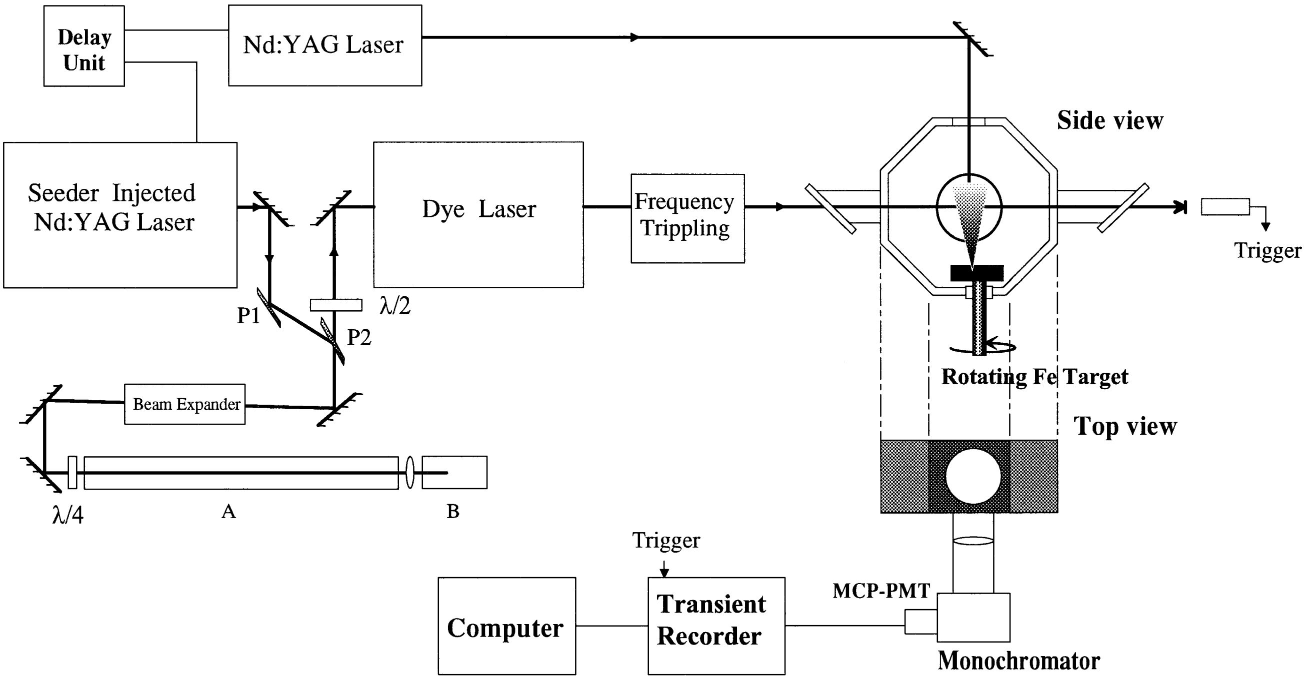

Fig. 1. Experimental set-up.

the first time on experimental radiative lifetimes using selective excitation of Fe II levels belonging to a higher subconfiguration, $3d^6(^3P)4p$. We have tested our technique on two levels, *viz.* $3d^6(^3P)4p\ z^4S^0_{3/2}$ and $3d^6(^3P)4p\ y^4P^0_{5/2}$, for which the lifetimes were measured using time-resolved laser induced fluorescence. A narrow bandwidth, short pulse excitation laser beam was obtained using a Stimulated Brillouin Scattering (SBS) compressor. Free Fe^+ ions with a sufficient population in the high metastable states were obtained in a laser-produced plasma.

2 Experimental set-up

Since Fe II is a complex spectrum with a very high spectral line density, a laser excitation pulse with a narrow bandwidth is needed for selective excitation of the level being studied. The lifetimes of the intermediate-excitation states were estimated to be around a few ns. For precision in the lifetime measurement a laser pulse with shorter duration is needed. In order to fulfil these requirements, an upgrade of the laser system based on the SBS in water was made, which enables the production of 1 ns laser pulses with a narrow bandwidth [14]. The experimental set-up is shown in Figure 1. The 532 nm beam of 8 ns pulses from a Nd:YAG laser (Continuum NY-82) is sent to the SBS compressor. Stimulated Brillouin scattering starts on the leading edge of the input pulse at the focus point in the water tube B (oscillator) as the laser power density reaches the threshold value. The phase conjugated reflection goes back along exactly the same way as the input beam and is amplified by the input beam during its passage through the water tube A (amplifier). The 8 ns input pulse is compressed to 1 ns in the reflection. The original vertically polarized beam changes its polarization direction to horizontal by the double passage through the $\lambda/4$-plate. The reflected 1 ns pulses go through the plate-polarizer P2, which is placed in the Brewster angle, and change to vertical polarization when passing through the $\lambda/2$-plate before being sent to pump the dye laser (Continuum Nd-60). Operating on the DCM dye, the output from the dye laser is tunable from 615 nm to 660 nm, with a pulse energy of 10-20 mJ and a pulse duration of about 1 ns. The bandwidth of the DCM red beam, measured with a pulsed wavelength-meter (Burleigh WA-4500) is less than 0.2 cm^{-1}. The dye laser output was frequency-tripled in a nonlinear optical crystal system (see detail in Ref. [15]), and the third harmonic was directed to cross the Fe^+ beam in a vacuum chamber.

Free Fe^+ ions with sufficient population in the high-lying metastable states were prepared in a laser-produced plasma. A 10 ns green laser pulse of about 10 mJ energy provided by a second Nd:YAG laser (Continuum Surelite) was focused perpendicularly onto a rotating iron target in the vacuum chamber. The expanding plasma was crossed by the excitation beam at a position of about 1 cm above the target. The delay time between the excitation and plasma-production pulses could be changed by externally triggering the two Nd:YAG lasers with a digital delay generator. Laser-induced fluorescence from the selectively excited Fe II level was collected by a fused-silica lens, appropriately filtered by a 1/8 m monochromator (resolution 6.4 nm/mm), and detected by a Hamamatsu 1564U microchannel-plate (MCP) photo multiplier (200 ps rise time). The data acquisition was performed by a digital transient recorder (Tektronix Model DSA 602) which had a 1 GHz

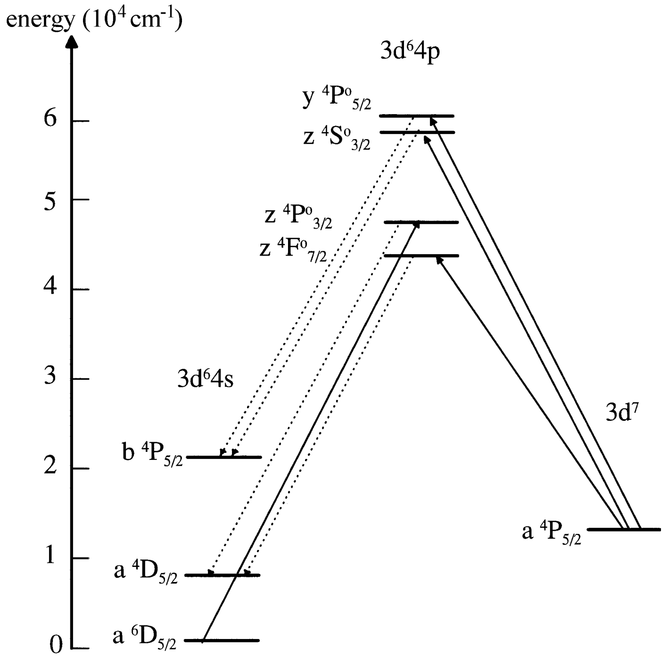

Fig. 2. Partial energy level diagram of Fe^+. The parent terms are indicated on the right of each level.

bandwidth and worked either in real time with a 2GS/s sampling rate or using interleaved sampling. The averaged time-resolved fluorescence decay curves were transferred to an IBM personal computer and lifetime evaluations were performed immediately.

3 Measurements and results

A partial energy level diagram of Fe II relevant to this experiment is shown in Figure 2. The low-lying metastable terms a^6D and a^4D belong to the $3d^64s$ configuration and have a ^5D term as their parent term, which means that, *e.g.*, $a^6D_{5/2} = 3d^6(^5D)4s\ ^6D_{5/2}$. Fine structure levels of these low terms can be used as the initial state to excite levels of the lowest odd-parity subconfiguration $3d^6(^5D)4p$ at 5 eV. In order to excite the z^4S^0 and the y^4P^0 levels of the higher subconfiguration $3d^6(^3P)4p$ at 7.5 eV, a sufficient population in the metastable ^4P levels of $3d^6(^3P)4s$ or $3d^7$ is required. As mentioned in the Introduction, the levels of $3d^6(^5D)4p$ have been thoroughly studied before, but no laser spectroscopic lifetime measurements of levels belonging to $3d^6(^3P)4p$ or higher subconfigurations have hitherto been published. The main barrier is to prepare an optically thin, collision free Fe^+ gas having a significant population in the high metastable states for Fe II. In the present experiment we have used a laser-produced plasma. Similar plasmas have previously been used to solve astrophysical spectroscopic problems by doing lifetime measurements of highly excited states in Pd II [16] and Zr II [17].

The plasma density and temperature in the observed region can be adjusted by changing the plasma production laser pulse energy, size of focus point, distance above

Table 1. Lifetimes measured in this work and comparisons with previous works.

Upper level	Energy (cm^{-1})	Excitation Lower level (cm^{-1})	λ_{voc} (nm)	Lifetimes this work (LIF)	Others
$z\ ^4P^0_{3/2}$	47389	$a\ ^6D_{7/2}(668)$	213.964	3.5(2)	3.44(11)[a]
$z\ ^4F^0_{7/2}$	44754	$a\ ^4P_{7/2}(13474)$	319.607	3.7(3)	3.63(11)[b]
					3.6(2)[b]
$z\ ^4S^0_{3/2}$	59663	$a\ ^4P_{7/2}(13474)$	216.434	3.8(2)	
$y\ ^4P^0_{5/2}$	60402	$a\ ^4P_{7/2}(13474)$	213.026	3.6(3)	

(^{a}) Reference [13].

(^{b}) Reference [10].

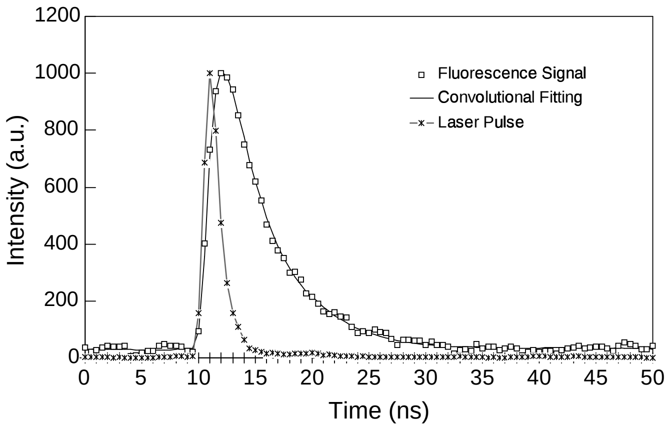

Fig. 3. A typical fluorescence signal from $z\ ^4S^0_{3/2}$, recorded excitation laser pulse and a convolution fit of the laser pulse with an exponential decay which has an inverted decay constant of $\tau = 3.8$ ns.

the target surface and delay time between the ablation and excitation pulses. As a first test, we measured the lifetime of the $z^4P^0_{3/2}$ state, which was excited from the low-lying metastable state $a^6D_{5/2}$ (668 cm^{-1}). This allowed us to use a cool plasma, and the delay time between the excitation and ablation beam can be varied from 0.4 to 2.3 μs. A consistent lifetime value within statistical fluctuations was obtained within this delay time. The two states around 60000 cm^{-1} were excited from the $a^4P_{5/2}$ level at 13474 cm^{-1}. In order to obtain a Fe+ beam with sufficient population in this high metastable level the plasma density and temperature had to be increased to magnitudes where effects of collisions and flight-out-of-view began to appear. By adjusting the parameters of the ablation beam, we found the measured lifetime values to be unaffected by collisions in a region in the front of the expanding plasma. That corresponded to delay times between 0.4 and 0.6 μs. Flight-out(or in)-of-view effects were checked by moving the monochromator so that the view position changed upwards and downwards around the excitation beam.

As a further test we measured the lifetime for the $z^4F^0_{7/2}$ state. This state can easily be reached from the ground using the third harmonic of a visible laser beam.

However, we used a longer wavelength and populated the state from the same metastable level and with the same plasma conditions as for the high-lying levels. As can be seen in Table 1, our value agrees with the ones of other investigators. To eliminate the saturation effect, the excitation pulses of different energy were used by inserting neutral density filters in the beam. Only weak signals were detected to keep the MCP working in the linear response region, and 2000 pulses were averaged to develop each curve. Laser pulses were recorded by the same detection system while putting in a metal rod to the laser beam in the interaction point. The lifetimes were evaluated by fitting the fluorescence signal to the convolution of the laser pulse with an exponential decay. A typical fit is shown in Figure 3. About 40 curves for each level were developed and the average value was taken as the final lifetime. The standard deviation and estimates of possible systematic errors were taken as the error bar.

4 Discussion

Spectral lines of Fe II are of great interest in astrophysics, directly or indirectly. The strength of an absorption line in a stellar spectrum is proportional to the population in the lower state and the oscillator strengths (f-values) of the transition. Under thermodynamical equilibrium conditions, this means a dependence of the product of $f \cdot \exp(-E/kT)$, the oscillator strength times the Boltzmann factor. Observed stellar Fe II lines can have a low value of this product, which is compensated by the high cosmic abundance of iron. For example, a 100 times larger abundance of iron than for another iron group element permits a 100 times smaller Boltzmann factor. A low value of the Boltzmann factor means high-lying metastable states.

Accurate oscillator strengths are therefore required for levels at intermediate and high excitation energies in Fe II. The critical point in getting experimental data is the measurement of radiative lifetimes, which are used in combination with branching fractions to get absolute f-values. The branching fractions are derived from emission line intensities, measured at a calibrated Fourier Transform Spectrometer (FTS). Also, accurate wavelengths are obtained using this instrument. Lines from different energy levels can be used to determine the iron abundance of a star and test the assumptions about thermal equilibrium. This is a direct use of the data which we plan to produce within the FERRUM project. The indirect use of our data concerns the numerous blends of Fe II and lines of more exotic elements, the analysis of which requires accurate Fe II data.

In the present investigation we have shown that laser spectroscopic techniques can be used for lifetime measurements of energy levels which do not combine strongly with the ground term. The laser-produced plasma is a source with high ion density and with metastable states fairly well populated. These levels can constitute plat forms for selective photon excitation and measurements of $3d^6 4p$ levels up to about 65000 cm^{-1}. The next step in the FERRUM project will be more lifetime measurements of $4p$ levels and also measurements of $4d$ states by two-step excitation. Simultaneously, measurements of branching fractions with the FTS will be performed.

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