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Beam-laser lifetime measurements for low-lying quartet states in Fe II

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Radiative lifetimes for seven levels of the $z\,^4P°$, $^4D°$, and $^4F°$ terms of Fe II have been measured by using laser excitation of a beam of fast Fe^+ ions. The ratio of the measured lifetime to that calculated from Kurucz's gf values appears to increase systematically with the energy of the levels involved.

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A recent analysis by Biémont *et al.* [1] of singly-ionized iron lines appearing in the solar spectrum has yielded a solar iron abundance 7.54±0.03 defined as $\log N_{\rm Fe}$ and taken relative to $\log N_{\rm H}$ being 12.00. This is in close agreement with the value found from chondritic meteorites, 7.51±0.01 [2], thus removing a discrepancy between previous estimates for the solar and meteoritic abundances. The analysis made use of some new beam-laser lifetime measurements to make an empirical correction to the theoretical gf values [3] used to obtain the iron abundance from the equivalent widths of 39 selected lines in the solar spectrum, this correction being about 12%. The lifetime measurements were for 12 levels of the $z\,^6P°$, $^6D°$, and $^6F°$ terms of Fe II. The mean ratio of the experimental and theoretical lifetimes was found to be 1.080±0.018 for the 5 $^6D°$ levels, whereas the mean ratio for the 2 $^6P°$ and 5 $^6F°$ levels was found to be 1.136±0.018. Since the $^6D°$ levels all have energies around 39 000 cm^{-1}, while the $^6P°$ and $^6F°$ levels lie 3000 to 4000 cm^{-1} higher, these results suggested a possible energy dependence for this empirical correction to the theoretical data. Furthermore, only 11 of the 39 solar transitions used in the abundance analysis have one of these sextets as their upper level. A further 23 of the solar transitions have one of the corresponding quartet levels as their upper state. We have therefore extended our beam-laser lifetime measurements to levels of the $z\,^4P°$, $^4D°$, and $^4F°$ terms in order to establish more exactly the empirical correction to the data calculated by Kurucz, and to investigate the possible energy dependence suggested by the sextet results. We have also measured the lifetime of the $z\,^6P^{\circ}_{3/2}$ level and remeasured the lifetime of the $z\,^6P^{\circ}_{5/2}$ level. In this report we discuss these new measurements and their implications for the solar iron abundance. Very recently, a new analysis by Hannaford *et al.* [4] has given a solar iron abundance of 7.48±0.04, in fair agreement with the value obtained by Biémont *et al.* In the present work, we also compare our lifetime results with those reported by Hannaford *et al.*

The lifetimes of the Fe II levels discussed in this report were all measured using laser-induced fluorescence (LIF) of a beam of Fe^+ ions from a 350-kV accelerator. The laser beam was arranged to intersect the ion beam at an angle of 45°. Measurements of the resulting Doppler shift were used to determine the ion velocity to better than ±1%. The fluorescence-decay curves were recorded by moving the detector system alternately parallel and antiparallel to the ion direction. The decay curves recorded in the two directions were compared to test for systematic variations associated with laser power or frequency drift. Three different laser dyes, Coumarin C460, C500, and C540A from Exciton, Inc., were used in these measurements. The dye laser output was frequency doubled by a β barium borate crystal to provide a tunable UV radiation from 230 to 275 nm with a mean power of 2–5 mW at a repetition rate of 250 Hz. Figure 1 shows the transitions used to populate the seven quartet levels studied in this work, using an excimer pumped EPD 330 dye laser from Lumonics.

Unlike the previous measurements for sextet states, which were all excited from the ground state, $a\,^6D$ multiplet, all our quartet measurements were excited from higher metastable states, $a\,^4F$ and $a\,^4D$. Most of the transitions were relatively weak due to the smaller ion population and hence the resonance peak signals were weaker than in our earlier experiments. It was therefore necessary to superimpose more sweeps of the decay curve in order to obtain adequate statistics for the lifetime measurements. This in turn required better laser power and pulse-to-pulse stability. As mentioned in our previous report [1], a simple feedback system was developed to monitor the laser power and change the discharge voltage of

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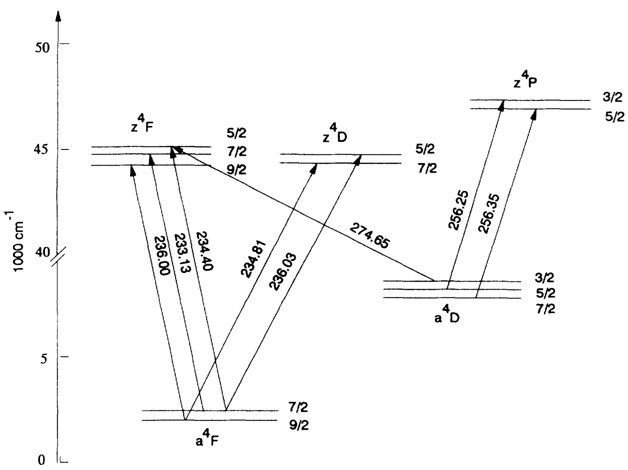

FIG. 1. Energy diagram showing the wavelengths (in nm) used to excite the quartet levels of Fe II studied here.

the excimer laser to stabilize the laser power. In the present work, a small fraction of UV radiation was also reflected onto a photodiode to monitor the energy of the laser pulse. A threshold was set to define a "good" laser pulse. If the number of good laser pulses dropped by more than 1%, the computer automatically repeated the data point. At least three decay curves were recorded for each level studied. Full details of our experimental procedures are available elsewhere [1,5].

The lifetimes determined from the fluorescence-decay curves for the seven quartet levels in Fe II studied here are listed in Table I, where they are compared with other experimental results, including the recent measurements reported by Hannaford *et al.* The agreement between the present data and those reported by Hannaford *et al.* using laser-induced fluorescence of a sputtered metal vapor is seen to be excellent, although the beam-laser technique being used here generally gives results of somewhat higher precision. Also included are the lifetimes derived from transition probabilities calculated by Kurucz, using the relativistic Hartree-Fock method coded by Cowan [6] and combined with a procedure to give a least-squares fitting to observed energy levels. The data currently available for the lifetimes of the $z\,^6P^\circ$ levels are summarized in Table II. In Fig. 2 we show the ratios of the beam-laser and calculated lifetimes as a function of the level energy for the 7 quartet and 13 sextet levels we have now studied. The presence of an apparently small but systematic increase in this ratio with energy is clear.

Figure 2 suggests that the $z\,^4D^\circ$ and $^4F^\circ$ levels give results that are not in agreement with the overall trend. It is therefore worth noting that the ratios obtained by summing the A values for transitions from the $z\,^4D^\circ$ and $z\,^4F^\circ$ states for the $j=\frac{5}{2}$ and $\frac{7}{2}$ levels in turn gives ratios that do lie on the overall trend. (The two points shown as solid circles in Fig. 2 represent the inverse of the ratios of the summed A values for each of the j values.) This is to be expected if the larger ratios observed for the $^4D^\circ$ levels are caused by an underestimate in the calculation of the interaction between the $^4D^\circ$ and $^4F^\circ$ states.

Given the apparent systematic trend shown in Fig. 2, it is natural to question its origin. Since all the wavelengths studied in the experiment lie in the narrow range between 233 and 275 nm, and all the levels have lifetimes in the range 3.0 to 3.9 ns, all the measurements have been conducted using essentially the same procedures. All the observed decays are followed for several lifetimes and, in all cases, the spatial resolution of the optical system corresponds to a time interval much less than the observed lifetime. Furthermore, tests using different ion energies revealed no dependence of the measured lifetimes on the ion velocity. We conclude that the systematic variation shown in Fig. 2 is more likely to have its origin in the calculations.

The consequences of our new lifetime measurements

TABLE I. Lifetimes of some quartet levels in Fe II.

States	J	Excitation wavelength (nm)	Lifetime (ns)			
			This work	HL[a]	HLGN[b]	Kurucz[c]
$z\,^4D$	$\frac{7}{2}$	234.81	3.02±0.07	3.7±0.4	3.1±0.2	2.43
	$\frac{5}{2}$	236.03	3.10±0.08	3.4±0.4	3.1±0.2	2.44
$z\,^4F$	$\frac{9}{2}$	236.00	3.87±0.09	4.1±0.3	3.7±0.2	3.34
	$\frac{7}{2}$	233.13	3.63±0.11	3.9±0.3	3.6±0.2	3.22
	$\frac{5}{2}$	234.40	3.75±0.14	4.0±0.3	3.7±0.2	3.26
		274.65				
$z\,^4P$	$\frac{5}{2}$	256.25	3.43±0.09	3.8±0.4		2.97
	$\frac{3}{2}$	256.35	3.44±0.11			2.96

[a]LIF measurement of Hannaford and Lowe (Ref. [9]).
[b]LIF measurement of Hannaford *et al.* (Ref. [4]).
[c]Obtained from multiconfiguration Hartree-Fock gf values calculated by Kurucz (Ref. [3]).

TABLE II. Lifetimes of the $z\,^6P^{\circ}$ levels in Fe II.

J	Excitation wavelength (nm)	Lifetime (ns)				
		This work	BBKAP[a]	HLGN[b]	SMH[c]	Kurucz[d]
$\frac{7}{2}$	234.35	3.73±0.06	3.73±0.05	3.8±0.2	3.5±0.3	3.28
$\frac{5}{2}$	233.38	3.79±0.12	3.83±0.07	3.7±0.2	3.5±0.3	3.26
	234.83					
$\frac{3}{2}$	233.80	3.71±0.12		3.6±0.2	3.4±0.3	3.25

[a]Fast beam-laser measurement of Biémont *et al.* (Ref. [1]).
[b]LIF measurement of Hannaford *et al.* (Ref. [4]).
[c]LIF measurement of Schade *et al.* (Ref. [10]).
[d]Obtained from multiconfiguration Hartree-Fock gf values calculated by Kurucz (Ref. [3]).

for the solar iron abundance are relatively minor, since the major source of uncertainty there is associated with the branching ratios required to obtain the gf values for the lines used in the solar analysis from the experimental lifetimes, rather than in the lifetimes themselves. Different approaches to this problem have been taken in the analysis by Biémont *et al.* [1] and by Hannaford *et al.* [4]. Biémont *et al.* assume the branching ratios calculated by Kurucz [3] and adjust the Kurucz gf values uniformly to give agreement with the experimental level lifetimes, whereas Hannaford *et al.* use experimental branching ratios having relatively large uncertainties. The Biémont *et al.* analysis, which gave an abundance of 7.54±0.03, relied on the lifetime measurements for 12 sextet levels, even though only 11 of the 39 solar lines used in the abundance analysis involved these levels. On the other hand, 23 of the solar lines involved levels belonging to the quartet terms discussed here. A correction of about 18% is more representative for these levels. Averaging over the quartet and sextet levels suggests that the abundance should be increased by 0.06, rather than by 0.05 as assumed previously, thus giving a final solar abundance of 7.55±0.03. However, Hannaford *et al.* [4] have pointed out that the systematic difference between the theoretical and experimental gf values shows a slight wavelength dependence, increasing slightly for shorter wavelength lines. Their solar analysis relies on experimental branching ratios by Heise and Kock [7] and Pauls, Grevesse, and Huber [8], renormalized by the lifetime measurements, to obtain the solar iron abundance from 15 solar lines in the Liège solar atlas [9]. The relatively low accuracy of the experimental branching ratios (typically of the order of ±20%) dominates the experimental uncertainties, with the result that repeating the Hannaford *et al.* analysis using our more precise lifetimes does not change the final iron abundance significantly from their estimate of 7.48±0.04. The weighted mean of the two analyses, including our lifetime

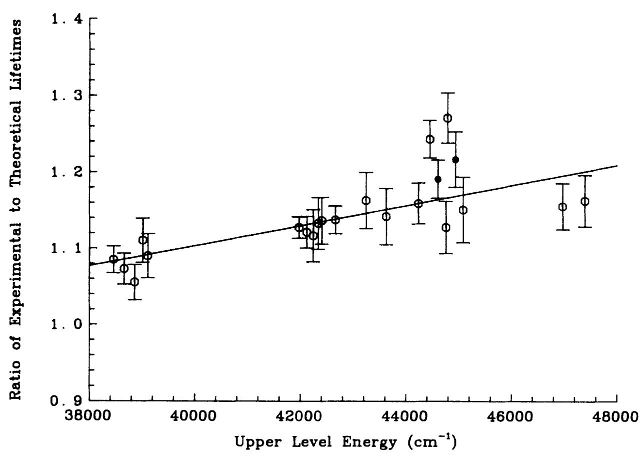

FIG. 2. Comparison of experimental and theoretical lifetimes as a function of the level energy. The ratio of experimental to theoretical lifetimes appears to increase with the energy. The two points with the solid circles (●) are the ratios obtained by summing the A values for transitions from $z\,^4D^{\circ}$ and $z\,^4F^{\circ}$ states for the $j=\frac{5}{2}$ and $\frac{7}{2}$ levels (see text). The slope of the least-squares-fitted line $(1.3\pm0.3)\times10^{-5}$ differs significantly from zero.

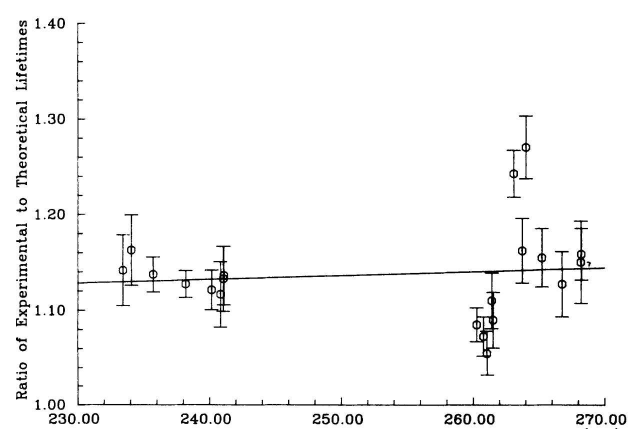

FIG. 3. Comparison of experimental and theoretical lifetimes as a function of the wavelength of the dominant decay transition (see text). The slope of the least-squares-fitted line $(3.9\pm9.0)\times10^{-4}$ is consistent with zero.

values, is then 7.52 ± 0.04, which we suggest is the best current estimate available for the solar iron abundance.

One final comment should perhaps be made regarding the empirical correction to the Kurucz calculations suggested by our results. Hannaford *et al.* [4] have pointed out that the difference between the experimental and calculated lifetimes for the Fe II levels under consideration implies that the theoretical gf values are systematically too high for the strong short-wavelength lines, which essentially determine the level lifetimes. They also draw attention to the small systematic increase in the solar iron abundance found by Biémont *et al.* [1] as the wavelength of the solar line used for the analysis is increased. Taken together, these observations suggest that there may be an overall wavelength dependence to the empirical correction to be applied to the Kurucz gf values. We have therefore searched for a possible systematic wavelength dependence in the ratio of the experimental and theoretical lifetimes for the Fe II levels we have studied. In most cases the lifetime is dominated by a single decay mode. For those cases where more than one transition contributes significantly to the decay of the given level, we have used a mean wavelength (weighted according to the relative A values) to represent that level. The ratio of the experimental and theoretical lifetimes of the various levels is plotted against the wavelength of the dominant decay branch(es) in Fig. 3, from which it is clear that there is in fact no significant dependence of the theoretical lifetimes on wavelength.

Note added in proof. An important contribution to the difference between the values obtained for the solar iron abundance by Hannaford *et al.* [4], and by Biémont *et al.* [1] and the present work, lies in the use of different solar chemical compositions to determine the electron and gas pressures in the solar photosphere. According to Hannaford *et al.* [4], applying their assumed composition to the analysis of Biémont *et al.* [1] would give an abundance of 7.51. Allowing for the change in the abundance of $+0.01$ found in the present work by including the lifetimes of the quartet levels would then give a final value of 7.52.

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