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New laboratory lifetime measurements of U II for the uranium cosmochronometer

H. Lundberg[1], S. Johansson[2], H. Nilsson[2], and Z. Zhang[1]

[1] Department of Physics, Lund Institute of Technology, PO Box 118, 22100 Lund, Sweden
[2] Department of Physics, Lund University, PO Box 118, 22100 Lund, Sweden
e-mail: sveneric.johansson@fysik.lu.se; hampus.nilsson@fysik.lu.se; zhang.zhiguo@fysik.lth.se

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Abstract. We present new measurements of radiative lifetimes for six energy levels of singly ionized uranium, U II, using laser-induced fluorescence technique. One of the levels, $5f^3 6d7p^6 M_{13/2}$ at 26191 cm^{-1}, decays by a transition at 3859.6 Å. This line has recently been observed in the spectrum of the metal-poor star CS1082-001, the first detection of uranium outside the solar system. The $\lambda 3859$ line can be used as the presently most accurate cosmochronometer (Cayrel et al. 2001). Our value of the lifetime of the $^6M_{13/2}$ level is 18.6 ± 0.7 ns, and it confirms the f-value used in the Nature article by Cayrel et al. (2001), which is based on an experimental lifetime of 20 ± 5 ns (Chen & Borzileri 1981). The new measurement also removes the doubt about the choice between that value and other f-values in the literature, differing by a factor of 3. Adopting the same branching fraction as Chen & Borzileri (1981) for the 3859.6 Å line, we derive a gf-value of 0.68, which is 8% higher than the value used by Cayrel et al. (2001). Of significance for the chronometer is also the reduced uncertainty of the radiative lifetime, 4% compared to 25%, and consequently of the f-value, which should decrease the uncertainty in the determination of the stellar age considerably.

Key words. atomic data – stars: evolution – Galaxy: evolution

1. Introduction

In a recent Letter in Nature, Cayrel et al. (2001) present the first detection of uranium outside the solar system by the identification of a U II line in the spectrum of the metal-poor star CS31082-001. The half-life of ^{238}U (4.5 Gyr) is about a third of the half-life of ^{232}Th (14 Gyr), and uranium is therefore more sensitive in dating the Galaxy than the previously used thorium chronometer. The dating is based on the "present" ratio of the relative abundances of a radioactive and a stable isotope, assuming that the ratio is known at the simultaneous production of the two elements. The dating problem is then reduced to the problem of determining the relative abundances, which is done by spectrum synthesis technique, where a computed spectrum is fitted to the observed spectrum. The computed spectrum depends on the validity of the stellar model atmosphere used and the atomic data for the relevant spectral lines. This Letter deals with atomic data for the U II line at 3859.6 Å observed and analysed by Cayrel et al. (2001), and radiative lifetimes for five more levels.

The wavelength of the 3859.6 Å line is accurate to better than 1 mÅ (Palmer et al. 1980), and it reflects the position of the ^{238}U isotope. The isotope shift ^{238}U$-^{235}$U might be considerable (Conway & Worden 1984), but is of no importance in this application, as all ^{235}U (half-life ≈ 0.7 Gyr) has decayed in the star.

The relative absorption depth of the U II line is determined by the abundance and the internal line strength, given by the oscillator strength (gf-value). Based on Voigt's arc measurements of U II lines (Voigt 1975), Corliss recalibrated his measurements of gf-values and produced a list of about 800 lines (Corliss 1976). Corliss used Voigt's gf-value of 0.24 for the 3859.6 Å line in his recalibration. The relative uncertainty in this value was estimated to be 5–10%, whereas the absolute uncertainty was 60%. The old value by Corliss was 0.58. Chen & Borzileri (1981) published a gA value of 2.8×10^8 s^{-1} for the 3859.6 Å line, which can be transferred to $gf = 0.63$. This value was obtained by applying a branching fraction (BF) of 0.403 to an experimental lifetime of 20 ± 5 ns, measured by laser induced-fluorescence technique. The BF was obtained from Fourier transform (FT) spectra and communicated privately to the authors. It is interesting to note that the Chen and Borzileri gf-value of 0.63 is close to the "old" Corliss value of 0.58, but far from the corrected Corliss value of 0.24, determined by Voigt.

Lifetime measurements of 25 U II levels were performed by Poulsen et al. (1981), but, due to the extended

Send offprint requests to: H. Lundberg,
e-mail: hans.lundberg@fysik.lth.se

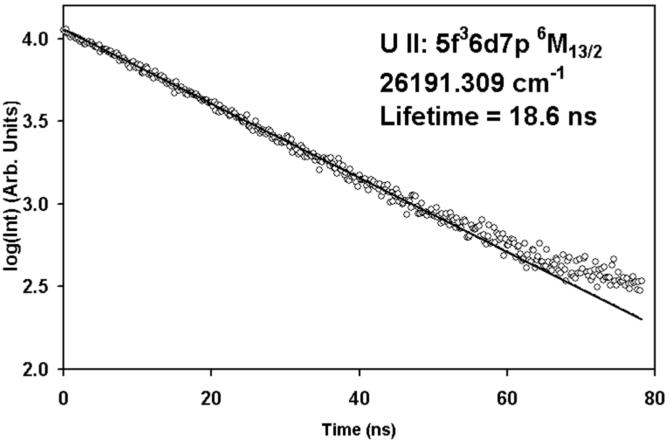

Fig. 1. Logarithmic decay curve of the $5f^36d7p^6M_{13/2}$ level of U II, observed after excitation at 4090.1 Å and detection of fluorescence at 3859.6 Å.

width of the laser excitation pulse, they could not measure the upper level of the 3859.6 Å transition, located at 26191 cm^{-1} (3.25 eV) above the ground level and labeled $5f^36d7p^6M_{13/2}$. The shortest lifetime measured by Poulsen et al. is 15 ns, and they estimated the lifetime of the $^6M_{13/2}$ level to be between 2 and 10 ns. This should be compared to the value of 20 ns reported by Chen & Borzileri (1981), which paper appeared 6 days after the paper by Poulsen et al. (1981) was received by the journal.

Two papers on gf-values in U II were published in 1987 by Compant La Fontaine (1987) and by Henrion et al. (1987). Compant La Fontaine (1987) combines published lifetimes and BFs to obtain gf-values, which means that he derives the same value (0.63) for λ 3859.6 Å as already reported by Chen & Borzileri (1981). Henrion et al. (1987) reports gf-values measured in a hollow-cathode plasma, for which the temperature is determined by means of gf-values from Palmer et al. (1980). This means that Henrion et al. (1987) get about the same low value as Palmer et al., 0.19 ± 0.013 and 0.23, respectively. Thus, there exists in the literature two sets of gf-values for the 3859.6 Å line, a low value at about 0.2 and a higher value at about 0.6, and Cayrel et al. (2001) chose the higher value in their new cosmochronometer.

In this paper we report on new lifetime measurements of six U II levels, including the level at 26191 cm^{-1} involved in the uranium cosmochronometer. For this level, we also discuss the BF in order to present an absolute gf-value. We have briefly looked at the BFs for transitions to the five other levels, but a more thorough investigation will be presented in a future paper.

2. Lifetime measurements

In the lifetime measurements uranium ions were produced by laser ablation. A solid target of UO_2 was irradiated with laser pulses and the measurements were performed about 1 cm from the target. Ions in the ablated plume were transferred to the level under investigation using pulsed tuneable laser radiation, produced by a Nd:YAG laser system. Conventional Nd:YAG laser pulses with a duration of about 10 ns were compressed to 1 ns utilizing stimulated Brillouin scattering in a water cell. The compressed pulses pumped a tuneable dye laser working on a red dye. The dye laser light was then converted to blue or ultraviolet wavelengths using frequency doubling and Raman shifting. The proper excitation wavelength for the studied level was set using the data of Palmer et al. (1980). Fluorescent light released at the decay of the excited levels was detected using a monochromator and a fast photomultiplier. A sampling oscilloscope with 1 GHz bandwidth performed the data acquisition. The experimental set-up is described in more detail in Li et al. (2000). To ensure a linear response of the detection system, only sufficiently weak fluorescence signals, subsequent to each excitation pulse, were sampled. An average of 1000–4000 pulses was necessary for each curve depending on the signal-to-noise ratio. Around 20 curves for each level were recorded and the averaged lifetime value was adopted as the final result. A decay curve is shown in Fig. 1. In the measurements precautions were taken to avoid effects of collisions, multiple scattering and flight-out-of-view effects.

The measured lifetime values are given in Table 1 and are compared with the results from the two earlier experimental investigations by Poulsen et al. (1981) and by Chen & Borzileri (1981). We also insert the estimates for two levels given by Poulsen et al. (1981). As we see in Table 1 all measurements agree within the error bars, which for our values and the data by Poulsen et al. (1981) are 4–6%, whereas those of Chen & Borzileri (1981) are 11% and 25% for two common data points. The error-bar of our lifetime values is an estimate of the combined random and systematic error. The standard deviations of different measurements are about 50% of the quoted uncertainties. The wavelengths in Table 1 refer to the transitions used for the excitation pulse and the fluorescence signal, respectively.

3. Branching fractions

The BF for the 3859.6 Å line was given by Chen & Borzileri (1981) as 0.403, based on laboratory spectra recorded with the FT spectrometer at Kitt Peak and privately communicated by J. Brault et al. The FT spectra are intensity calibrated and give a direct possibility to determine the BF, if all lines are observed. The three strongest lines from the upper level, $^6M_{13/2}$, in the 4000 Å region are present in the atlas of the FT spectrum by Palmer et al. (1980), and they give a BF of 0.50 for the λ3859 line, and 0.40 and 0.10 for the two other lines. The same three decay channels were observed in our lifetime measurements, and the relative intensities support the BF values derived from Palmer et al. (1980). However, there are 9 more lines from the $^6M_{13/2}$ level present in the grating spectrum recorded at Los Alamos (Steinhaus et al. 1971). These spectra are recorded on photographic plates,

Table 1. Experimental radiative lifetimes of six U II levels.

Config.	Term	J	Energy cm^{-1}	Excitation λ(Å)	Fluorescence λ (Å)	Exp. lifetime (ns) This work	Poulsen et al.	Chen et al.
$5f^3 7s7p$	^6K	4.5	23315.090	4341.7	4341.7	84(5)	85(4)	
$5f^3 7s7p$	^6I	4.5	24684.132	4050.0	4050.0	49(3)		44(5)
–	–	6.5	25714.049	4171.6	3932.0	35(2)	34(2)	
$5f^3 6d7p$	^6M	6.5	26191.309	4090.1	3859.6	18.6(7)	2-10[a]	20(5)
$5f^3 6d7p$	^6L	5.5	28154.450	3550.8	3865.9	12.4(6)	2-10[a]	
$5f^3 6d7p$	^6M	7.5	30341.675	3496.4	3881.5	24.0(1.5)		

[a] Estimate given by Poulsen et al. (1981).

and are therefore not easily intensity calibrated. The FT spectrum in the atlas by Palmer et al. (1980) was recorded at Kitt Peak from a hollow cathode lamp and run at a direct current of 70 mA. The archive at Kitt Peak also contains FT spectra of uranium recorded at a higher current, 300 mA. We have studied these spectra and found ten lines from the $^6M_{13/2}$ level, thereby verifying the BF reported in the paper by Chen & Borzileri (1981). Thus, from this BF of 0.403 and the new radiative lifetime of the $^6M_{13/2}$ level of 18.6 ns, we can derive a gf-value of 0.68. Since Chen & Borzileri (1981) do not give any uncertainty of the BF it is not possible to assign an uncertainty to the gf-value.

However, new measurements of relative intensities using the Lund FT spectrometer are in progress in order to solve this problem.

4. Conclusion

By measuring the radiative lifetime of the $5f^3 6d7p\,^6M_{13/2}$ at 26191 cm^{-1} of U II and applying a previously reported BF for the λ3859.6 transition, we can derive a gf-value of 0.68 for this cosmologically important line. The U II line has been observed in the spectrum of a metal-poor star and provides, together with a number of thorium lines, the presently most accurate cosmo-chronometer (Cayrel et al. 2001). The small uncertainty (about 4%) in the new lifetime measurements allows for a much smaller uncertainty in the radioactive dating of stars.

The new measurements have also solved the problem with two sets of gf-values in the literature for the λ3859.6 line, differing by a factor of 3. The unknown uncertainty of the BF for the line has motivated us to remeasure the relevant line intensities in the U II spectrum, a work in progress at the Lund Fourier Transform spectrometer. The BF data for the $5f^3 6d7p\,^6M_{13/2}$ level and for the other five levels, for which lifetimes have been reported in this work, will be the subject for a coming paper. Work is also in progress on gf-values for Th II lines, relevant for the cosmochronometer (B. Nordström, private communication).

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