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Lifetime measurements for doubly ionized uranium

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Abstract

The first lifetime measurements for doubly ionized uranium are reported in the present work. They relate to five levels belonging to the configurations $5f^26d^2$, $5f^26d7s$ and $5f^37p$ and have been obtained with a time-resolved laser-induced-fluorescence technique, the ions being produced by laser ablation of a uranium oxide target.

1. Introduction

Up to now, there have been no radiative data at all (transition probabilities or lifetimes) available for U III. The reasons is that U III ions are not easily produced in the laboratory. In addition, the complexity of the configurations involved in the excited states and the fragmentary knowledge of the spectrum of this ion have prevented theoretical calculations.

Transition probabilities for U III transitions however are expected to be important in astrophysics, and particularly in cosmochronology where the age of a star can be determined by the use of a radioactive isotope of sufficiently long lifetime. ^{238}U , which represents 99.3% of the whole uranium content of the solar system, is the longest-lived uranium isotope among the 13 isotopes and isomers and has a half-life of 4.47 Gyr. It was pointed out recently by Goriely and Clerbaux (1999) that new accurate observations of heavy radioactive elements could put the Th cosmochronometry on a firmer footing, especially if *Th and U lines could be observed simultaneously* in metal-poor stars, as already pointed out by others (Arnould and Takahashi 1990).

Up to now, no U III transitions, however, have been considered in cosmochronology, the main reason being the complete lack of radiative transition probabilities for this ion. As a first step in that direction, we report in the present work on the first lifetime measurements for U III. A first set of transition probabilities for Th III have been reported very recently (Biémont *et al* 2002).

Table 1. U III levels measured, corresponding excitation schemes and experimental lifetime values.

Measured state ^a	Level energy (cm ⁻¹)	Lower level (cm ⁻¹)	Excitation		$\lambda_{\text{obs(air)}}$ (nm)	τ (ns)
			$\lambda_{\text{exc(air)}}$ (nm)	Scheme ^b		
5f ² 6d ² 5I ₄	28 922.088	885.332	356.572	2ν + S	356.6	104 ± 10
5f ² 6d ² , J = 4	31 469.442	885.332	326.873	2ν	326.9	41 ± 3
5f ² 6d7s, J = 5	33 236.715	210.265	302.700	2ν	351.9	150 ± 15
5f ³ 7p 5K ₅	36 651.404	885.332	279.512	2ν + AS	304.8	23 ± 2
5f ² 6d7s J = 4	37 559.544	885.332	272.590	2ν + AS	304.4	44 ± 3

^a From Blaise and Wyart (1992).^b The meaning of the symbols is as follows: 2ν: frequency doubling; AS: first-order anti-Stokes component; and S: first-order Stokes component.

2. Experimental set-up used for the lifetime measurements

Radiative lifetime measurements for five levels of U III, belonging to the 5f²6d², 5f²6d7s and 5f³7p configurations, have been carried out using a time-resolved laser-induced-fluorescence technique. In the experiment, free U III ions have been produced by employing laser ablation of a uranium oxide target. Selective excitation was realized by using a tunable UV laser pulse with a short pulse duration. The experimental schemes used in the measurements are summarized in table 1.

The experimental set-up used in the experiments is similar to the one described in previous papers (see e.g. Zhang *et al* (2001)). Pulses from a frequency-doubled Nd:YAG laser (Continuum Surelite), usually with laser pulse energy in the range of 2–10 mJ, were focused vertically onto the surface of a compressed uranium oxide tablet target, which was rotated by a direct-current motor. After the pulses, the plasma containing uranium atoms and ions in different ionized stages expanded from the target. In order to obtain short-duration pulses to excite the ions measured, the radiation from an injection-seeded and frequency-doubled Nd:YAG laser (Continuum NY-82) was sent to a stimulated Brillouin scattering (SBS) pulse compressor. The output from the SBS compressor, with about 1 ns pulse duration and a single-pulse energy of about 200 mJ, was utilized to pump a dye laser (Continuum Nd-60), employing DCM or R610 + R640 dye according to the different excitation requirements. The properly tuned dye laser output was frequency doubled in a KDP crystal. The second-harmonic radiation from the crystal was used to excite the ions from a metastable state to two of the levels studied. The other three levels investigated were excited using the first-order Stokes or anti-Stokes stimulated Raman scattering (SRS) component derived from the second-harmonic radiation. The Stokes or anti-Stokes component was produced by focusing the second-harmonic radiation into a hydrogen cell with a gas pressure of 10 bar. The excitation laser beam was selected using a quartz Pellin–Broca prism and it was focused by two quartz lenses to the centre of a vacuum chamber, where it interacted with the ions about 10 mm above the target.

The two Nd:YAG lasers were triggered externally by a digital delay generator (Stanford Research System Model 535), which was also used to adjust a delay time between the ablation and excitation pulses. When the U²⁺ ions in the plasma reached the interaction zone, 10 mm above the foil, the ions were excited by the excitation laser beam passing through the plasma horizontally by properly choosing trigger parameters of the two lasers. In the measurements, the fluorescence from the level measured was imaged by two CaF₂ lenses onto the entrance slit of a vacuum monochromator, and was detected by a Hamamatsu R3809U-58 photomultiplier. The time-resolved signal was acquired and averaged, employing a digital transient recorder

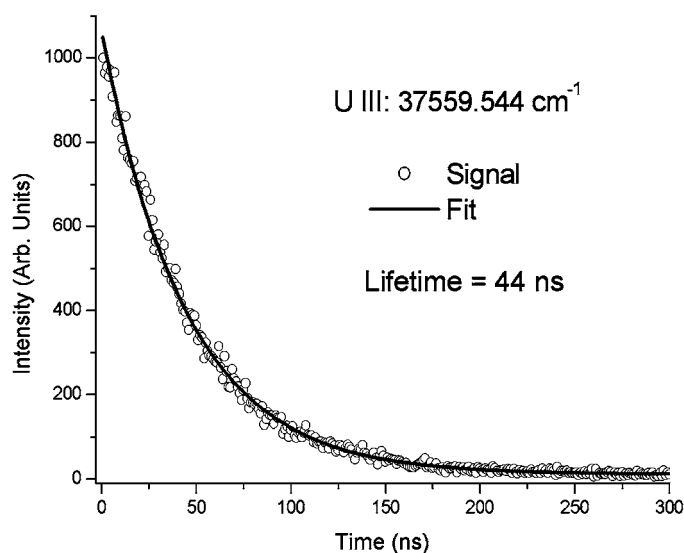


Figure 1. A typical experimental time-resolved fluorescence signal from the level at $37\,559.544\text{ cm}^{-1}$ in U III. The lifetime deduced from the fit is $44 \pm 3\text{ ns}$.

(Tektronix Model DSA 602), and the averaged time-resolved fluorescence decay curve was sent to a personal computer for lifetime evaluation.

Before the measurements, it was carefully verified that there were no identified lines for the elements U and O, belonging to neutral atoms and ions in different ionization stages in the spectral neighbourhood of the excitation wavelength. In the measurements, it was further verified that the observed fluorescence signal was emitted from U^{2+} ions by checking at which delay time the fluorescence signal was maximum (the speeds of different species are different). In order to collect the fluorescence effectively, the entrance slit of the monochromator was placed horizontally and it was opened maximally to avoid flight-out-of-view effects. However, this was not sufficient to fully avoid flight-out-of-view effects when the maximum fluorescence signal was selected, because such a signal was built up mostly from ions with a high speed. The delay time was thus actively increased in order to interact with the low-speed tail of the U^{2+} ions, clearly at the expense of intensity. When the time-resolved fluorescence decay curve was recorded, a suitable magnetic field of about 60 G was added in the plasma zone by a pair of Helmholtz coils in order to reduce by spatial separation the recombination between the electrons and the excited ions.

In order to get a reasonable signal-to-noise ratio, each decay curve was obtained by averaging fluorescence photons from more than 4000 pulses. During the experiments, a variety of experimental conditions were changed, including the delay time, the intensity of the excitation laser and that of the ablation laser. This resulted in a change of the fluorescence signal intensity by a factor of 5. However, it was found that the lifetime values had no clear systematic trend. The lifetime evaluation was performed using an exponential fit. Seven to ten curves were recorded for each level; a typical curve is seen in figure 1. The final lifetime result for each level was taken by the averaging of the data for the curves.

3. Results and discussion

The five lifetimes deduced are shown in the last column of table 1, where the error bars reflect not only the statistical errors, but also a conservative estimate for possible remaining systematic errors. As a small scatter was observed among the different results obtained for each level under different experimental conditions, indicating that collisional effects as well as radiation trapping were probably negligible, it was decided to adopt for the error bars of all the levels except the long-lived ones the uncertainties resulting directly from this scatter. For the two levels with lifetimes larger than 100 ns, the errors were increased by about 5% to take into account the eventual larger sensitivity to collisional effects. There are no data, either theoretical or experimental, available for comparison.

Twelve transitions depopulating the levels measured in the present work are quoted in the list of U III transitions observed in the laboratory, most of them being rather strong (Blaise and Wyart 1992). Obtaining transition probabilities from the lifetime measurements requires knowledge of adequate branching fractions. In the present case, the use of theoretical methods for deducing such data is prevented by the complexity of the spectrum considered. More precisely, it appears that the branching fractions deduced from the use of a pseudo-relativistic Hartree–Fock (HFR) approach (Cowan 1981), taking configuration interaction and the most important relativistic effects into account, were not reliable. In fact, a model retaining as interacting configurations $5f^4$, $5f^37p$, $5f^26d7s$, $5f^26d^2$, $5f^27s^2$, $5f^27p^2$ for even parity and $5f^36d$, $5f^37d$, $5f^37s$, $5f^26d7p$, $5f^27s7p$, $5f6d^3$, $5f6d^27s$ and $5f6d7s^2$ for odd parity shows that a large number of transitions depopulating the levels of interest are substantially affected by cancellation effects perturbing the calculation of the line strengths. As there is no strongly dominating decay transition for the levels measured, the theoretical transition probabilities are likely to be strongly dependent upon such effects and, consequently, are expected to be unreliable. In addition, it was verified that the introduction in the calculations of core-polarization (CP) effects, using a CP potential (Migdalek and Baylis 1978) (with the parameters $\alpha_d = 9.78 a_0^3$ and $r_c = 1.383 a_0$, these values corresponding to the dipole polarizability of the ionic core UV (Fraga *et al* 1976) and to the expectation value of r for the outermost core orbitals $6p^6$), and a correction to the dipole matrix elements similar to that considered with success recently for different complex atomic systems (see e.g. Biémont *et al* (2001), Li *et al* (2001)) does not improve upon the situation. Accurate laboratory measurements of branching fractions are urgently needed to provide the required transition probabilities.

The first lifetime measurements for doubly ionized uranium reported in the present work, together with similar data obtained very recently for doubly ionized thorium, open the way to the use of U III and Th III transitions as cosmochronometers for hot Ap stars. It is expected that these new accurate laser measurements, combined with the necessary experimental branching fractions as soon as they become available, will allow a quantitative analysis of the high-resolution line profiles now currently observed in metal-poor Ap stars.

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