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Time-resolved laser-induced fluorescence measurements of Rydberg states in Lu I and comparison with theory

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Abstract

Time-resolved laser-induced fluorescence measurements have been performed for ten odd Rydberg states of neutral lutetium, belonging to the $6s^2(^1S)np$ ($n = 8-9$) and $6s^2(^1S)nf$ ($n = 5-8$) series. For $6s^2(^1S)8p$ and $6s^2(^1S)7f$, the experimental lifetimes corresponding to the two J values within the doublet differ substantially. Comparison with theoretical values, calculated with extensive configuration interaction and core-polarization effects included, shows that the experimental trends are adequately reproduced both for np ($n = 8-9$) and nf ($n = 5-8$) states.

1. Introduction

Lutetium ($Z = 71$) is the heaviest atom in the group of the lanthanides. ^{175}Lu is by far (97%) the dominant of the two stable isotopes present in the solar system, ^{176}Lu representing only 3%. Lu has been identified in the solar photosphere, and its abundance, which is one of the lowest of the rare-earth group, is deduced from two Lu II lines, located at 339.706 and 622.187 nm. The last revision of the solar lutetium abundance ($A_{\text{Lu}} = 0.06 \pm 0.07$, in the usual logarithmic scale, where the hydrogen abundance is equal to 12.00) is due to Bord *et al* (1998), and is in good agreement with the meteoritic result, $A_{\text{Lu}} = 0.12 \pm 0.01$, according to the compilation of Anders and Grevesse (1989). This result, based on theoretical f -values, needs to be assessed by accurate experimental results.

Despite its low cosmic abundance, lutetium has been found in different Ap stars in the form of Lu I or Lu II (see e.g. Jaschek and Brandt 1972 or Poli *et al* 1987), and the analysis of the stellar spectra requires accurate atomic data, the final aim being an improved knowledge

of the chemical composition of the different types of stars and, *in fine*, input to the model of star formation.

The analysis of the high-resolution stellar spectra requires information about the atomic transition probabilities but also about the hyperfine structure (HFS) constants (Kurucz 1993), both isotopes of Lu showing HFS patterns (the nuclear spin being $I = 7/2$). The HFS and isotope effects in neutral lutetium have been investigated by different groups in the past: in particular, data have been published by Lin *et al* (1994), Reddy and Rao (1989), Kuhnert *et al* (1983) and Göbel (1970a, 1971).

Experimental lifetimes along the Rydberg series, particularly for higher-energy states, are a useful test of the theoretical models, the configuration interaction becoming increasingly complex when the myriad of close-lying high-energy levels is involved.

Transition probabilities or lifetimes in singly ionized lutetium have been the subject of a limited number of experimental or theoretical investigations in the 1970s: beam-foil spectroscopy analyses were published by Andersen and Sorensen (1974) and by Andersen *et al* (1975). More recently, experimental work based on selective laser excitation has been published by Den Hartog *et al* (1998) and by Fedchak *et al* (2000). On the theoretical side, a multiconfiguration Dirac-Fock study of the $6s^2\ ^1S_0$ - $6s6p\ ^3P_1^o$, $^1P_1^o$ transitions in the Yb isoelectronic sequence has been reported by Migdalek and Baylis (1987). Time-resolved laser-induced fluorescence studies in doubly ionized lutetium are also due to Biémont *et al* (1999) and to Fedchak *et al* (2000). Dipole polarizability and core-polarization effects have been studied by the same authors and by Migdalek (1980, 1982).

The transition probabilities in neutral lutetium are more poorly known and are still extremely scarce, despite the fact that some efforts have been devoted, during the last 30 years, to the investigation of this atom. A review paper on the resonance transitions was published by Doidge (1995a). The compilation of the Lu I lifetimes published by Blagoev and Komarovskii (1994) contains 42 values, but they concern only the levels of the configurations $5d6s6p$, $6s^2$ and $6s^2nd$ ($n \geq 19$). A double-resonance investigation of excited states is due to Göbel (1970b). Laser spectroscopic studies were reported by Kwiatkowski *et al* (1980), and Rydberg states were studied by laser multistep resonance ionization spectroscopy (Vidolova-Angelova *et al* 1992). Lifetime measurements in Lu I are due to Gorshkov *et al* (1984) and Vidolova-Angelova (1992), but the most detailed and recent analysis has been published by Fedchak *et al* (2000), who performed time-resolved laser-induced fluorescence measurements on a slow beam of lutetium atoms. In the latter case, 22 odd-parity levels and four even-parity levels were measured.

For the different reasons stated above, we have decided to perform a new experimental investigation of some Rydberg states of neutral lutetium (i.e. the $6s^2(^1S)np$ ($n = 8-9$) and $6s^2(^1S)nf$ ($n = 5-8$) states), using a laser-induced fluorescence technique. The measurements were compared to calculations performed with a relativistic Hartree-Fock (HFR) method (Cowan 1981) in order to assess the reliability of the theoretical model.

2. The atomic structure of neutral lutetium

The atomic spectrum of neutral lutetium, although partly based on old studies, is fairly well known (Martin *et al* 1978). In fact the NIST compilation is firstly based on levels deduced by Meggers and Scribner (1930, 1937), who made arc and spark observations in the 250–1077 nm region. Extensions of the analysis are due to Klinckenberg (1954) and Camus and Tomkins (1972), who made observations in the 228–370 nm region. Additional contributions are due to Pinnington (1963).

Neutral lutetium has a rather simple electronic structure, with three electrons outside a closed shell, formed by the 14 f electrons. It appears, therefore, as an interesting case for

Table 1. Lu I levels measured and the corresponding excitation schemes.

State ^a	Lower level (cm ⁻¹)	Upper level (cm ⁻¹)	Excitation	Scheme ^b λ_{vac} (nm)	Detection λ_{vac}
6s ² (¹ S)8p ² P _{1/2} ^o	0.0	36 808.76	271.67	2 ω + AS	271.7
6s ² (¹ S)8p ² P _{3/2} ^o	1993.92	37 131.38	284.59	2 ω + AS	284.6
6s ² (¹ S)9p ² P _{3/2} ^o	1993.92	39 424.68	267.15	3 ω + 2S	267.2
6s ² (¹ S)5f ² F _{5/2} ^o	1993.92	36 633.31	288.68	2 ω + AS	273.0
6s ² (¹ S)5f ² F _{7/2} ^o	1993.92	36 644.12	288.59	2 ω + AS	288.6
6s ² (¹ S)6f ² F _{5/2} ^o	0.0	39 212.61	255.02	3 ω + 2S	255.0
6s ² (¹ S)6f ² F _{7/2} ^o	1993.92	39 220.17	268.62	3 ω + 2S	268.6
6s ² (¹ S)7f ² F _{5/2} ^o	0.0	40 626.82	246.14	3 ω + 2S	246.1
6s ² (¹ S)7f ² F _{7/2} ^o	1993.92	40 619.01	258.89	3 ω + 2S	258.9
6s ² (¹ S)8f ² F _{5/2} ^o	0.0	41 456.26	241.21	3 ω + S	241.2

^a From Martin *et al* (1978).

^b 2 ω means frequency doubling; 3 ω means frequency tripling; AS, S and 2S stand for the anti-Stokes and the first- and the second-order Stokes components, respectively.

testing the accuracy of the theoretical models. The identified even and odd configurations are 5d6s², 5d²6s, 6s²ns ($n = 7-14$), 6s²nd ($n = 6-34$), 5d6s7s and 6s²np ($n = 6-27$), 5d6s6p, 5d²6p and 6s²nf ($n = 5-35$), respectively.

The NIST energy levels (Martin *et al* 1978) were used in the fitting procedure of the present work (see section 3).

3. Lifetime measurements

The lifetimes of nine levels of Lu I were measured in the present work, with the time-resolved laser-induced fluorescence technique. One-step excitation was used to excite these states due to the opposite parity of the ground configuration 5d6s². The levels of interest have been excited according to the schemes summarized in table 1.

The experimental set-up used in our experiment is illustrated in figure 1. Lu atoms were produced in a laser-induced plasma, using 532 nm wavelength laser pulses emitted from a 10 Hz repetition rate and 10 ns duration Nd:YAG laser (Continuum Surelite), with variable pulse energy. Pulse energies in the range 2–10 mJ were normally used. The pulses were focused on the surface of a Lu foil, rotating in a vacuum chamber. In order to obtain the required excitation, another Q -switched and seeded injection Nd:YAG laser (Continuum NY-82), with a pulse duration of 8 ns, was combined with a stimulated Brillouin scattering compressor to shorten the pulses down to 1 ns. The laser was used to pump a dye laser (Continuum Nd-60), in which a DCM dye was used in the experiments. According to the excitation requirements, the second- or third-order harmonics of the dye laser were obtained employing a non-linear optical system, including a KDP crystal, a retarding plate and a BBO crystal. The measured levels were excited by the different Raman-shifted components of the harmonics, obtained in a cell with hydrogen at 10 bar, when the harmonics were focused into the cell. In the experiments, the excitation light was isolated with a prism and focused by a lens at a distance of about 1 cm above the foil. The two Nd:YAG lasers were triggered by a digital delay generator (Stanford Research System, model 535), and the delay between the ablation and excitation pulses was adjusted by the generator. The atoms produced by the ablation pulse were excited selectively

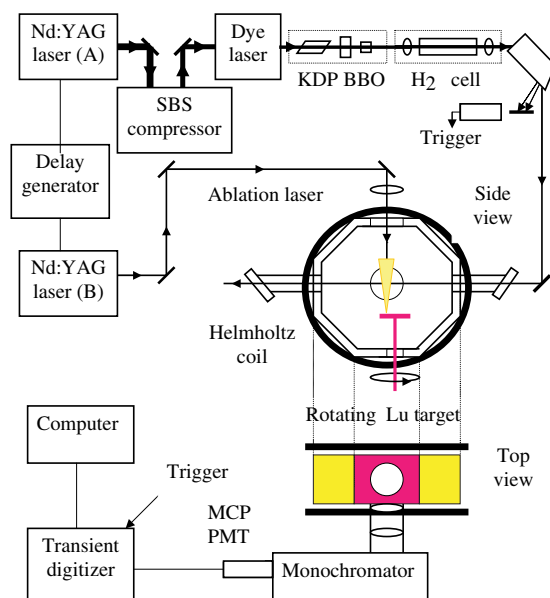


Figure 1. Experimental set-up used for the lifetime measurements.
(This figure is in colour only in the electronic version)

by the excitation laser beam crossing the plasma horizontally. The fluorescence, emitted from the excited levels, was imaged by a fused-silica lens and focused on the entrance slit of a 1/8 m monochromator. A Hamamatsu 1564U micro-channel plate was used for the detection.

The time-resolved signal was recorded and averaged with a digital transient recorder (Tektronix model DSA 602), and the fluorescence decay curve was sent to a personal computer for lifetime determination. The lifetime evaluation was performed by an exponential fit.

In order to be sure that the Lu I transitions of interest were indeed studied, the modification of the fluorescence signal as a function of the delay time was investigated, the occurrence of a maximum fluorescence signal appearing at longer delay times for Lu I than for various Lu ions. This is because atoms have considerably lower speeds than ions in their flight from the ablation site. The lower speed makes atoms less vulnerable to so-called flight-out-of-view effects. If the detection efficiency is altered due to changing geometry during the signal observation time interval, a pure exponential decay will not be observed. By using a wide monochromator slit and optimizing its geometrical position, detrimental effects can be eliminated.

In order to obtain a sufficiently high signal-to-noise ratio, a decay curve was obtained by averaging fluorescence photons from more than 2000 pulses.

For each level measured, more than seven fluorescence decay curves were recorded under different experimental conditions, the delay time and intensities of the ablation and excitation laser pulses being varied. Thus, the measurements were performed for different concentrations and temperatures of the plasma. The evaluated lifetimes from the different curves were found to agree well. This showed that there were no saturation, reabsorption or collisional effects occurring, and the final result was formed by averaging the lifetime values. The nine lifetimes measured are reported in the third column of table 2. The error bars reflect the statistical scattering, but also a conservative estimate of the possible remaining systematic errors, adjusted in such a way that an overall accuracy of about 10% is stated for each lifetime studied.

Table 2. Theoretical and experimental lifetime values in Lu I, and comparison with previous results. (c)–(e) HFR calculations with a scaling factor of 0.85, 0.60 and 0.50, respectively, for the R^k integrals between $6s^2np$ ($n = 8-9$) and $5d6s6p$.

Levels ^a	Upper level (cm ⁻¹)	Experimental lifetimes (ns)		Theoretical lifetimes (ns)		
		This experiment	Previous experiments	(c)	(d)	(e)
$6s^2(1S)7p\ 2P_{1/2}^o$	29 430.90		9.2(5) ^b , 8.6(8) ^c	8.1	8.3	8.4
$6s^2(1S)7p\ 2P_{3/2}^o$	30 488.62		5.9(3) ^b , 7.4(6) ^c	6.3	6.5	6.6
$6s^2(1S)8p\ 2P_{1/2}^o$	36 808.76	44(4)		22	35	46
$6s^2(1S)8p\ 2P_{3/2}^o$	37 131.38	20.5(2.0)		9	12	15
$6s^2(1S)9p\ 2P_{1/2}^o$	39 321.96			82	142	178
$6s^2(1S)9p\ 2P_{3/2}^o$	39 424.68	82(10)		44	84	113
$6s^2(1S)10p\ 2P_{1/2}^o$	40 661.02			138	142	143
$6s^2(1S)10p\ 2P_{3/2}^o$	40 735.33			89	95	96
$6s^2(1S)5f\ 2F_{5/2}^o$	36 633.31	33.1(3.0)	30.2(1.5) ^b , 31(2) ^c	30	30	30
$6s^2(1S)5f\ 2F_{7/2}^o$	36 644.12	30.7(3.0)	30.3(1.5) ^b , 29(2) ^c	32	32	32
$6s^2(1S)6f\ 2F_{5/2}^o$	39 212.61	52(5)		52	52	52
$6s^2(1S)6f\ 2F_{7/2}^o$	39 220.17	50(5)		56	56	56
$6s^2(1S)7f\ 2F_{5/2}^o$	40 626.82	54(5)		68	68	68
$6s^2(1S)7f\ 2F_{7/2}^o$	40 619.01	66(7)		71	71	71
$6s^2(1S)8f\ 2F_{5/2}^o$	41 456.26	104(10)		127	127	127
$6s^2(1S)8f\ 2F_{7/2}^o$	41 460.08			133	133	133
$6s^2(1S)9f\ 2F_{5/2}^o$	42 000.72			83	83	83
$6s^2(1S)9f\ 2F_{7/2}^o$	42 006.00			87	87	87

^a From Martin *et al* (1978).

^b Fedchak *et al* (2000).

^c Gorshkov *et al* (1984).

4. The calculations

The theoretical HFR approach used was described previously by Fedchak *et al* (2000). Configuration interaction and core-polarization effects were taken into account in a detailed way. Configuration interaction was considered among the configurations $5d6s^2$, $6s^2ns$ ($n = 7-10$), $6s^2nd$ ($n = 6-10$), $6s^2ng$ ($n = 5-10$), $6s6p^2$, $5d^2ns$ ($n = 6-10$), $5d^2nd$ ($n = 6-10$), $5d6sns$ ($n = 7-10$), $5d6snd$ ($n = 6-10$), $5d6p^2$ and $5d^3$ (even parity) and $6s^2np$ ($n = 6-10$), $6s^2nf$ ($n = 5-10$), $5d^2np$ ($n = 6-10$), $5d^2nf$ ($n = 5-10$), $5d6snp$ ($n = 6-10$) and $5d6snf$ ($n = 5-10$) (odd parity). These configuration lists extend considerably those considered in our previous work (Fedchak *et al* 2000). More precisely, we have added the even-parity configurations $6s^2ng$ ($n = 5-10$), $5d^2ns$ ($n = 8-10$), $5d^2nd$ ($n = 8-10$), $5d6sns$ ($n = 8-10$) and $5d6snd$ ($n = 8-10$) and the odd-parity configurations $5d^2np$ ($n = 8-10$), $5d^2nf$ ($n = 8-10$), $5d6snp$ ($n = 8-10$) and $5d6snf$ ($n = 8-10$) in order to better represent the interaction along the $6s^2ns$, $6s^2np$, $6s^2nd$ and $6s^2nf$ Rydberg series up to $n = 10$.

The core-polarization effects were considered in the same way as in the HFR(A) approach of Fedchak *et al* (2000), since this model was found to lead to a very good agreement with the experimental lifetimes in the cases of $6s^27s$, $6s^27p$ and $6s^25f$ levels. More precisely, the core-polarization potential and the correction to the dipole operator, as described by Quinet *et al* (1999), were included in the model using the dipole polarizability of the ionic core, α_d ,

Table 3. Variation of the $6s^28p$ and $6s^29p$ radiative lifetimes, as a function of the eigenvector compositions (only two components are quoted).

SF ^a	Configuration	<i>J</i>	Composition (%)	Lifetime (ns)
0.85	$6s^28p$	1/2	88.5 $8p^2P$ + 4.6 $5d6s(^3D)6p^2P$	21.6
	$6s^28p$	3/2	76.5 $8p^2P$ + 11.6 $5d6s(^3D)6p^2P$	8.9
	$6s^29p$	1/2	94.7 $9p^2P$ + 0.8 $5d6s(^3D)6p^2P$	82.1
	$6s^29p$	3/2	93.2 $9p^2P$ + 1.7 $5d6s(^3D)6p^2P$	44.2
0.75	$6s^28p$	1/2	90.2 $8p^2P$ + 3.7 $5d6s(^3D)6p^2P$	25.5
	$6s^28p$	3/2	79.4 $8p^2P$ + 10.0 $5d6s(^3D)6p^2P$	9.8
	$6s^29p$	1/2	95.1 $9p^2P$ + 0.6 $5d6s(^3D)6p^2P$	101.6
	$6s^29p$	3/2	94.0 $9p^2P$ + 1.3 $5d6s(^3D)6p^2P$	56.4
0.65	$6s^28p$	1/2	91.7 $8p^2P$ + 2.8 $5d6s(^3D)6p^2P$	31.1
	$6s^28p$	3/2	82.3 $8p^2P$ + 8.3 $5d6s(^3D)6p^2P$	11.2
	$6s^29p$	1/2	95.5 $9p^2P$ + 0.4 $5d6s(^3D)6p^2P$	126.8
	$6s^29p$	3/2	94.7 $9p^2P$ + 0.9 $5d6s(^3D)6p^2P$	73.3
0.60	$6s^28p$	1/2	92.4 $8p^2P$ + 2.4 $5d6s(^3D)6p^2P$	34.9
	$6s^28p$	3/2	83.8 $8p^2P$ + 7.4 $5d6s(^3D)6p^2P$	12.2
	$6s^29p$	1/2	95.7 $9p^2P$ + 0.3 $5d6s(^3D)6p^2P$	142.0
	$6s^29p$	3/2	95.0 $9p^2P$ + 0.8 $5d6s(^3D)6p^2P$	84.2
0.50	$6s^28p$	1/2	93.7 $8p^2P$ + 1.6 $5d6s(^3D)6p^2P$	45.7
	$6s^28p$	3/2	87.0 $8p^2P$ + 5.6 $5d6s(^3D)6p^2P$	15.2
	$6s^29p$	1/2	95.9 $9p^2P$ + 0.2 $5d6s(^3D)6p^2P$	178.3
	$6s^29p$	3/2	95.5 $9p^2P$ + 0.5 $5d6s(^3D)6p^2P$	112.9

^a Scaling factor applied to the configuration interaction integrals, R^k , between the $6s^2np$ ($n = 8-9$) and $5d6s6p$ configurations.

equal to $5.20 a_0^3$, as tabulated by Fraga *et al* (1976) for the Lu IV ion, and a cut-off radius, r_c , equal to $1.406 a_0$, which corresponds to the expectation value of r for the outermost core orbital ($5p^6$), as calculated with Cowan's codes.

Using a well established least-squares fitting procedure, the radial parameter values were adjusted to obtain the best agreement between the calculated and the experimental energy levels taken from Vergès and Wyart (1978) and from the NIST compilation (Martin *et al* 1978). The Slater integrals (F^k , G^k and R^k), not optimized semi-empirically, were scaled down by a factor equal to 0.85 (see, however, the discussion of section 5).

5. Results and discussion

The calculated lifetimes are given in the fifth, sixth and seventh columns of table 2, where they are compared with the experimental results obtained in the present work and with previous experimental data. The three sets of results, (c), (d) and (e), correspond to different scaling factors applied to some configuration interaction integrals (see the notes to table 2). When comparing these values (calculation (c)) with the laser measurements of the present work, a rather good agreement (from 0 to 30%) is observed along the $6s^2nf$ ($n = 6-8$) Rydberg series. The calculated values remain a factor of two smaller than the experimental lifetimes for the $6s^2np$ ($n = 8, 9$) levels. It is interesting to observe that the ratio between theory (calculation (c)) and experiment (this work) is nearly constant along the np series, and that the fine-structure variation of the lifetimes is correctly reproduced. Looking into the details, it appears that the calculation of radiative lifetimes of the np ($n = 8, 9$) levels is very sensitive to the mixing with the $5d6s(^3D)6p^2P$ states. Indeed, when slightly changing the admixture of

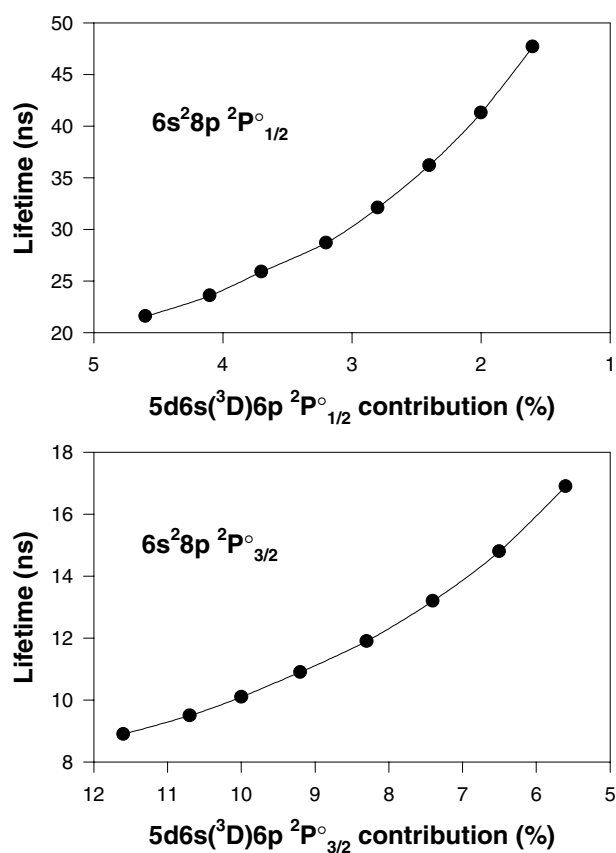


Figure 2. Behaviour of the lifetimes of $8p \ ^2P^{\circ}_{1/2}$ and $8p \ ^2P^{\circ}_{3/2}$ levels as a function of the mixing with the $5d6s(^3D)6p \ ^2P^{\circ}$ contribution.

this latter state in the eigenvector compositions of $6s^2 8p$ and $6s^2 9p$ levels (simply by changing the scaling factor applied to the R^k integrals connecting these configurations, the cases (c), (d) and (e) corresponding to scaling factors of 0.85, 0.60 and 0.50, respectively), it appears that the calculated lifetimes for $8p$ and $9p$ are considerably modified, this variation even reaching a factor of two when the scaling factor is varied from 0.85 to 0.50. This corresponds in fact to a change of a few per cent in the $6s^2 8p$ and $6s^2 9p$ purities. The present discussion is clearly illustrated in table 3 and in figure 2, which show the variation of the lifetimes as a function of the eigenvector compositions.

It should be emphasized that the $6s^2(^1S)nf \ ^2F^{\circ}_{5/2,7/2}$ levels are not dependent upon the choice of the scaling factor, as shown in table 2. The lifetimes of the $5d6s(^3D)6p \ ^2P^{\circ}$ levels, previously measured by Fedchak *et al* (2000), are much less sensitive than the $5d6s(^3D)np \ ^2P^{\circ}$ levels ($n = 8, 9$) to the choice of this scaling factor: indeed, using 0.50 instead of 0.85 would lead to $\tau = 6.0$ and 3.0 ns for the levels $5d6s(^3D)6p \ ^2P^{\circ}_{1/2}$ ($32\,058.10 \text{ cm}^{-1}$) and $5d6s(^3D)6p \ ^2P^{\circ}_{3/2}$ ($34\,436.49 \text{ cm}^{-1}$), values which are compatible with the published experimental results, $\tau = 4.9(0.2)$ and $3.7(0.2)$ ns, respectively.

An experimental lifetime of the $9p \ ^2P^{\circ}_{1/2}$ state would also have been very valuable in view of the above considerations. However, in spite of a thorough search, laser-induced fluorescence could not be detected in the present experiments.

The theoretical lifetimes of the present work are also compared in table 2 with previous experimental measurements for the 7p and 5f levels, due to Fedchak *et al* (2000) and to Gorshkov *et al* (1984). The agreement observed is good for the four levels, the HFR result being located in between the two experimental results for 7p $^2P_{3/2}^o$.

Considering the experimental lifetimes and the theoretical branching fractions as obtained in the present work, it has been possible to deduce oscillator strengths for the transitions depopulating the levels of interest. In view of space limitations, the detailed results are not presented here. The extensive tables will be available in the database of astrophysical interest, DREAM, which is progressively created at the address <http://www.umh.ac.be/~astro/dream.shtml>.

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