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Lifetime measurements of odd-parity high-excitation levels of Sm II by time-resolved laser spectroscopy

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ABSTRACT

Natural radiative lifetimes of 53 odd-parity highly excited levels of singly ionized samarium in the energy range 26 599.08–36 107.66 cm^{-1} have been measured by the time-resolved laser-induced fluorescence technique in an ionic plume produced by laser ablation. The fundamental frequency and second harmonic of a dye laser have been adopted as the tunable exciting source (308–650 nm), and a fast detection system has been used to record fluorescence signals. The lifetime values obtained in this paper are in the range 10.5–271 ns. A good agreement between the present results and the previously published values was achieved. This work will be useful for investigating the composition of chemically peculiar stars.

Key words: atomic data – atomic processes – methods: laboratory – techniques: spectroscopic.

1 INTRODUCTION

Singly ionized samarium, which is characterized by the 4f^{6}6s ground configuration, is one of the rare-earth elements. The radiative data of Sm II are of great interest in different fields of physics. In astrophysics, emission lines of Sm II have been identified in the spectra of some chemically peculiar stars such as the Ap stars of the Cr–Eu–Sr subgroup (Aikman, Cowley & Crosswhite 1979), Bp stars (Cowley & Crosswhite 1978), Ba stars (Danziger 1965; Lambert 1985) and the S-type stars (Bidelman 1953). In particular, the three transitions at 456.6, 471.9 and 673.1 nm appear in the spectral types F5 to M2 (Gopka & Komarov 1990). Moreover, Sm II has also been found to be present in the extreme peculiar star HD 101065 (Przybylski's star) with a confidence level = 95 per cent (Cowley & Mathys 1998; Cowley et al. 2000). It is well known that the combination of accurate radiative lifetimes with precise branching fractions will provide the best experimental determination of the absolute transition probabilities, these quantities being crucial parameters in the determination of solar and stellar abundances (Whaling et al. 1985). So, the interest in radiative lifetimes of the ionized rare-earth elements has rapidly increased. On early investigations of the lifetimes of Sm II, only a few results of low-lying odd-parity levels have been measured using different techniques including beam-foil spectroscopy, the delayed-coincidence technique with electron-beam excitation and beam-laser method by various authors (Andersen et al. 1975; Blagoev et al. 1978; Gorshkov & Komarovskii 1986; Vogel et al. 1988). Employing the laser-induced fluorescence (LIF) technique skillfully, the extensive investigation production of radiative lifetimes of Sm II was obtained. Using the LIF technique, the lifetimes of 35 levels up to 30 880 cm^{-1} and those of 82 levels up to 29 600 cm^{-1} were measured by Biémont et al. (1989) and Scholl et al. (2002), respectively. Soon afterwards, the radiative lifetimes of 47 levels over the energy range 21 000–36 000 cm^{-1} were measured by Xu et al. (2003). Recently, with the same technique, radiative lifetimes of 212 odd-parity levels up to 38 505.66 cm^{-1} were measured by Lawler et al. (2006). Moreover, some compilation works of lifetimes and f-values on Sm II have been accomplished by various authors (Blagoev & Komarovskii 1994; Morton 2000). However, due to the complexity of this ion originating from the large number of closely spaced low-excitation levels, the lifetimes of many odd-parity high-excitation levels with a principal emitting spectrum in the ultraviolet region, which should be dominant in stellar spectra, have not been investigated adequately. In addition, sufficient ionic data, such as the energy levels, natural radiative lifetimes as well as the transition probability, should be helpful to gain an insight into the properties of highly excited levels of ions with different charges.

For all the above reasons, the investigation of radiative lifetimes concerning Sm II is necessary and timely. In this experiment, the

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natural radiative lifetimes of 53 odd-parity highly excited levels of Sm II have been investigated using the time-resolved LIF technique in an ionic plume of laser-produced plasma.

2 EXPERIMENTAL SETUP

The experimental setup used here for lifetime measurements is shown in Fig. 1. In this experiment, the plasma generated through laser ablation on a samarium sample with a purity of 99.99 per cent has been employed as a stable free ion source. Previously, this technique has proved very efficient and reliable for lifetime measurements (Li et al. 2000; Zhang et al. 2010a,b). A 5-mJ, 532-nm pulse, with about 8 ns duration, emitted by a Q-switched Nd:YAG laser (Continuum Precision II) working at a 10-Hz repetition rate, was focused on a rotary thin target plate. The fundamental radiation and its second harmonic of a linearly polarized dye laser (Sirah Cobra-Stretch), operating with DCM dyes, which was pumped by a Q-switched Nd:YAG 355 nm laser (Spectra-Physics Quanta-Ray Pro-Series) working at 10 Hz with about 8 ns pulse duration, were employed as tunable excitation radiation from 308 to 650 nm. The linewidth of the dye laser was about 0.08 cm^{-1}. Then, the excitation laser with about 5–7 ns pulse duration was sent horizontally through the vacuum chamber, where it intersected the vertical ionic plume about 10 mm above the target. By a digital delay generator (Stanford Research Systems Model 535), the delay between excitation and ablation pulses can be arbitrarily adjusted. Following the excitation, the fluorescence signal was focused on a grating monochromator ($f = 10$ cm) by a fused silica lens in the direction perpendicular to the laser and the ionic plume, and then it was detected by a photomultiplier tube (PMT) (Hamamatsu R3896). In order to enhance the fluorescence collection efficiency, the monochromator was rotated by an angle of 90° so that its entrance slit was horizontal and parallel to the excitation beam and, in this case, the entrance slit had to be properly widened to avoid the flight-out-of-view effect. In order to register the time-resolved fluorescence photocurrent signal from the PMT, a 500-MHz digital oscilloscope (Tektronix TDS 620B) triggered by the excitation laser with a fast photoelectric diode was connected with a computer through a GPIB cable to record and store signal data.

In the direction parallel to the horizontal component of the Earth's magnetic field, an appropriate magnetic field of about 130 G produced by a pair of Helmholtz coils was used to wash out the quantum beats produced by the Earth magnetic field (Den Hartog et al. 2005), and it is found that the applied magnetic field can effectively reduce the recombination background from the plasma (Zhang et al. 2001). Besides, a proper magnetic field generated by another pair of coils was employed to neutralize the vertical component of the Earth's field.

3 LIFETIME MEASUREMENTS

In the measurements, to ensure that only one state is studied at a time, the wavelength of the excitation light for each state was chosen from the available excitation paths in order to avoid the co-excitation of other levels. For the Sm II levels under study, the ultraviolet transition channels with larger branch fractions were generally used for excitation paths, while other smaller branches in the visible region were used for the fluorescence detection for

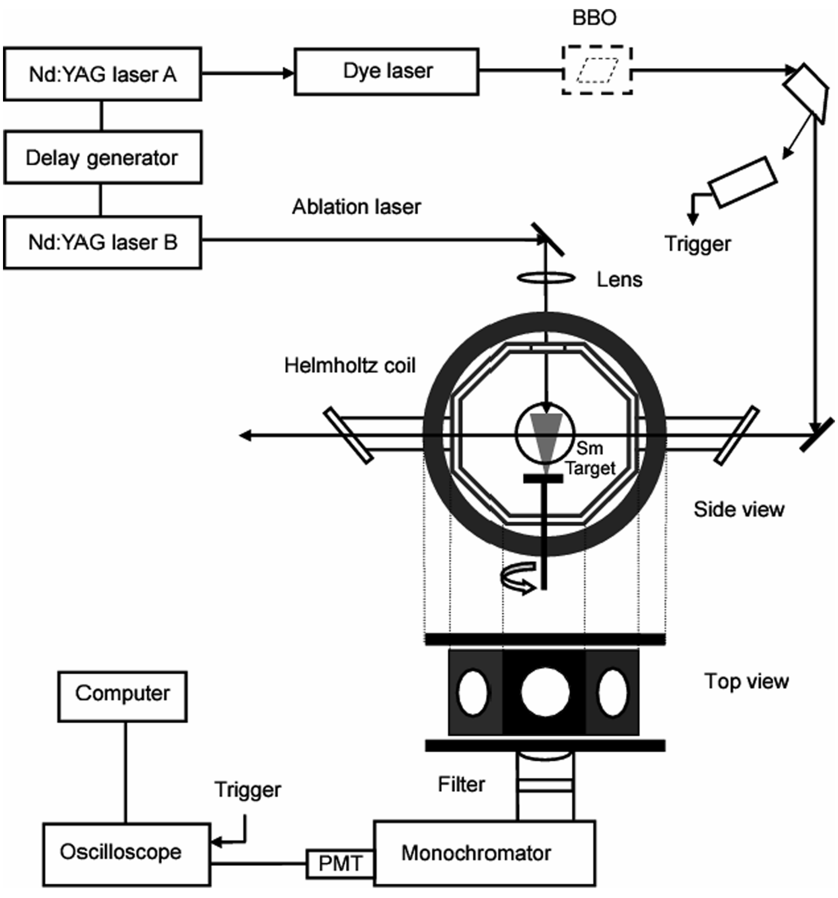

Figure 1. Experimental setup used for lifetime measurements.

avoiding the effect of stray light from the excitation laser. The excitations of selected levels have been confirmed by checking the disappearance of the signal when blocking the ablation laser and verifying that all the fluorescence channels were related to this level.

By changing the ablation pulse energy, the size of the focus point, the distance between the target surface and the excitation pulse, the delay time and the intensity of the excitation pulses, and the plasma conditions, such as particle speed, density and temperature as well as the density of excited ions in the excitation region, could be effectively adjusted (Zhang et al. 2000). Under the appropriate plasma and excitation conditions, with the delays between 5 and 50 μs, the intensities of the detected fluorescence signals were changed by a factor of 5–10, and the evaluated lifetimes remained constant within the experimental errors. This indicated that the possible effects of flight-out-of-view, radiation trapping, super-radiation, PMT non-linear response and collision-induced quenching have been effectively checked and eliminated in this process. For the ionic levels with lifetimes longer than 100 ns, more caution has been taken for avoiding the effect of flight out of view caused by ionic motion. For the lifetime measurements of such levels, the delay time was set longer than 10 μs with which the atoms in the excitation region travelled 10 mm; the atomic speed was known to be about 1000 m s^{-1}. To observe the entire decay process of a level, an acquisition time of five times the radiative lifetime is necessary. For the longest lifetime of about 270 ns in this paper, Sm ions flew about 2.7 mm in the vertical direction during the observation time. Considering that the diameter of about 2 mm of the exciting laser beam and the imaging character of our fluorescence collection system, a slit width of about 5 mm was used in the experiment. With this slit width and properly increasing delay time, the flight-out-of-view effect can be effectively reduced. Also due to the large band width, at the selected observation wavelength in the visible region, several florescence channels of an excited level could be synchronously detected by the PMT. This will bring on higher florescence collection efficiency than the case of only one channel. Moreover, through changing the pressure in the vacuum chamber from 2×10^{-4} to 3×10^{-3} Pa, the possible collision with remnant gas was checked, while the flight-out-of-view effects were considered again by changing the position of the monochromator slit. In addition, the effects of radiation trapping, non-linear response of the PMT detector and super-radiation have been analysed and eliminated by modifying the pulse energies of the ablation and excitation lasers when the delay between the excitation and ablation pulses was appropriate. Finally, the black-body radiation (BBR) effect (Theodosiou 1984) could be negligible in this work since each investigated level not only has large energy spacing with respect to the adjacent levels accessible by a dipolar transition, but also has a much shorter lifetime compared with the BBR relaxation time.

For improving the signal-to-noise ratio, more than 2000 shots were averaged for each fluorescence decay curve. For a level with a lifetime longer than 80 ns, which corresponds to 10 times the excitation pulse duration, the lifetime value may be evaluated by a least-squares exponential fitting to the recorded fluorescence curves. In this process, to make sure that no deviations from an exponential shape of the signal influences the measurement, the starting point of the fitting procedure has to be chosen cautiously. A typical fluorescence decay curve of the 33 772.54 cm^{-1} level with an exponential fitting for lifetime evaluation is shown in Fig. 2. For the levels with shorter lifetimes, a deconvolution of the fluorescence decay curves is necessary to take into account the effects of the finite duration of the excitation laser pulse and the limited response time of the detection system (Mayo et al. 2006). In this case, besides the fluorescence

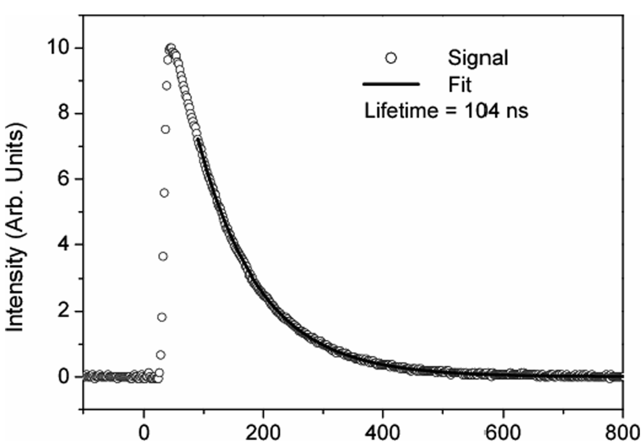

Figure 2. Typical fluorescence decay curve of the 33 772.54 cm^{-1} level with an exponential fitting for lifetime evaluation.

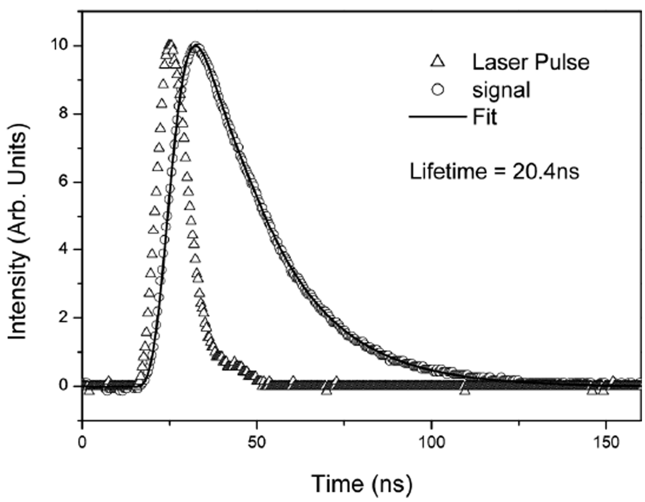

Figure 3. Fluorescence decay curve of the 34 133.81 cm^{-1} level together with the fitted convolution curve between the laser pulse and an exponential with a decay constant of 20.4 ns.

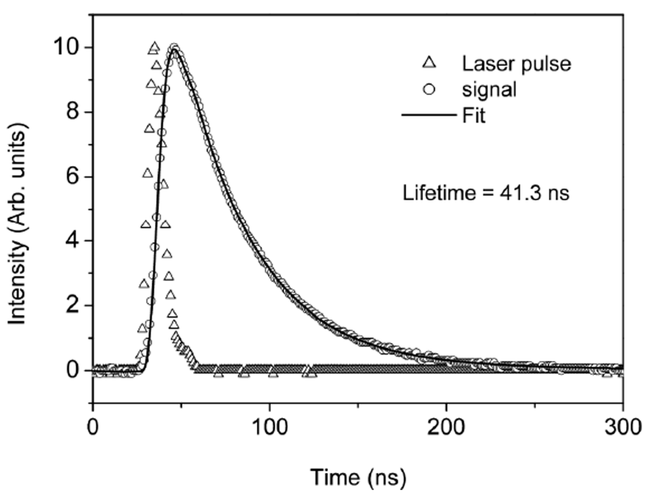

Figure 4. Typical fluorescence decay curve of the 32 945.19 cm^{-1} level together with a convolution fitting and with a decay constant of 41.3 ns.

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Table 1. Measured lifetimes for odd-parity levels of Sm II and comparison with previous results. The excitation (Exc.) and fluorescence (Obs.) wavelengths are also given.

Upper level		Origin level		Exc.	Obs.	Lifetime (ns)	
E (cm^{-1})[a]	J[a]	E (cm^{-1})[a]	J[a]	(nm)	(nm)	This work	Previous
26 599.08	3/2	10 518.50	3/2	621.868	406.6	19.0(0.9)	17.9(2)[b], 20.0(1.0)[c], 17.9[d]
28 997.14	9/2	13 466.50	11/2	643.888	363.5	10.5(0.6)	10.1[e], 9.62(15)[f], 12(2)[g], 9.6(5)[b], 9.7[d]
30 252.90	1/2	0	1/2	330.547	530.3	11.4(0.7)	10.6[d]
31 052.45	3/2	0	1/2	322.036	515.7	98.2(2.3)	98.3[d]
31 186.00	1/2	326.64	3/2	324.051	537.1	43.2(1.0)	
31 309.40	3/2	0, 326.64	1/2, 3/2	319.397, 322.760	508.9	32.9(1.9)	32.1[d]
31 599.63	5/2	326.64	3/2	319.765	505.0	50.8(1.0)	51.4[d]
31 638.79	3/2	326.64	3/2	319.365	573.2	58.9(2.5)	
31 725.56	5/2	326.64	3/2	318.482	491.9	17.8(0.9)	16.9[d]
31 774.52	7/2	838.22	5/2	323.245	555.6	27.7(1.2)	25(2)[c], 23.8[d]
31 915.67	7/2	1489.16	7/2	328.661	551.3	26.8(1.3)	29(2)[c], 26.6[d]
31 954.10	3/2	326.64	3/2	316.181	466.5	162(4.0)	
32 067.40	1/2	0	1/2	311.843	512.8	22.6(2.0)	20.4[d]
32 223.45	3/2	838.22	5/2	318.621	554.6	60.7(3.0)	
32 397.48	5/2	326.64, 1489.16	3/2, 7/2	311.810, 323.537	495.9	78.7(2.4)	80[d]
32 492.65	3/2	326.64, 838.22	3/2, 5/2	310.887, 315.912	468.7	98.4(4.0)	
32 549.64	7/2	1489.16	7/2	321.953	478.7	24.0(0.9)	23.1[d]
32 603.65	5/2	326.64	3/2	309.818	557.5	30.8(1.0)	
32 685.70	5/2	1489.16	7/2	320.548	528.9	243(4.0)	
32 945.19	9/2	1489.16	7/2	317.904	627.4	41.3(1.5)	42(3)[c], 40.1[d]
33 218.75	5/2	1489.16	7/2	315.163	539.1	78.0(1.7)	
33 227.90	3/2	838.22	5/2	308.740	525.4	20.0(0.8)	
33 252.45	9/2	1489.16	7/2	314.829	615.5	132 (4.1)	
33 286.30	11/2	2237.97	9/2	322.078	508.1	61.5(1.8)	61.0[d]
33 539.60	5/2	2003.23	3/2	317.094	584.4	45.7(2.1)	
33 576.60	7/2	2237.97	9/2	319.095	528.9	48.0(1.0)	
33 598.70	11/2	2237.91	9/2	318.870	512.4	32.4(0.7)	33(2)[c], 31.2[d]
33 613.43	9/2	2237.97	9/2	318.720	484.8	168 (10)	
33 630.20	7/2	2688.69	5/2	323.190	543.9	26.1(1.2)	
33 689.50	5/2	2003.23	3/2	315.594	562.1	271(6.0)	
33 772.54	9/2	2237.97	9/2	317.112	576.6	104(3.0)	
33 775.84	11/2	3052.65	11/2	326.487	559.3	29.7(1.5)	35(2)[c], 32.1[d]
33 809.85	13/2	3909.62	13/2	334.446	581.6	73.5(7.0)	80[d]
33 852.93	9/2	2237.97	9/2	316.306	537.3	35.9(1.9)	
34 066.75	5/2	2003.23	3/2	311.881	555.9	71.9(2.3)	69.5[d]
34 068.41	3/2	2003.23	3/2	311.865	503.1	20.2(0.9)	
34 133.81	11/2	2237.97	9/2	313.521	499.5	20.4(0.6)	
34 145.44	13/2	3052.65	11/2	321.618	468.3	16.0(0.5)	16.0(0.9)[c], 14.6[d]
34 205.91	11/2	2237.97	9/2	312.813	568.5	62.3(1.0)	
34 418.95	11/2	3052.65	11/2	318.814	521.5	40.6(2.4)	38(2)[c], 35.5[d]
34 453.83	5/2	2003.23	3/2	308.161	592.2	52.2(0.6)	
34 505.80	7/2	2688.69	5/2	314.296	464.7	64.1(2.0)	
34 722.26	9/2	3499.12	7/2	320.275	444.6	138(4.0)	
34 745.47	13/2	3052.65, 3909.62	11/2, 13/2	315.529, 324.298	494.0	122(3.0)	119[d]
34 890.85	13/2	3909.62	13/2	322.776	480.6	62.5(1.5)	62.4[d]
34 951.90	9/2	3499.12	7/2	317.937	479.9	82.2(3.0)	
35 101.70	7/2	2688.69, 3499.12	5/2, 7/2	308.516, 316.430	586.5	75.8(2.0)	
35 192.21	11/2	3909.62	13/2	319.667	518.3	20.8(0.6)	
35 261.20	15/2	3909.62	13/2	318.963	430.7	208 (12)	
35 303.60	7/2	4386.03	9/2	323.441	546.5	143(2.0)	
35 463.91	9/2	3052.65	11/2	308.535	511.1	58.4(1.0)	
35 583.40	5/2	3499.12	7/2	311.679	512.7	49.9(2.0)	
36 107.66	9/2	4386.03	9/2	315.242	441.7	140(4.8)	

[a] From Martin, Zalubas & Hagan (1978).
[b] From Scholl et al. (2002).
[c] From Xu et al. (2003).
[d] From Lawler et al. (2006).
[e] From Biémont et al. (1989).
[f] From Vogel et al. (1988).
[g] From Blagoev et al. (1978).

signal, the exciting laser pulse, which corresponds to the product of the real intensity of the laser pulse and the instrumental response function, needs to be recorded by the same detection system alternately. The typical fluorescence decay curve of the 34 133.81 cm^{-1} level, together with the fitted convolution curve of the laser pulse and an exponential with a 20.4 ns decay lifetime, is shown in Fig. 3. In addition, the fluorescence decay curve of the 32 945.19 cm^{-1} level, of which the radiative lifetime has also been measured by Lawler et al., together with a convolution procedure, is shown in Fig. 4.

In the least-squares exponential fittings, for each fluorescence curve, two lifetime values were determined with two starting points. One, with the strongest intensity, was chosen at a position outside the region affected by the stray light of the excitation laser and the other was at a position with half the intensity of the former point. In the convolution procedure also two lifetime values were evaluated using two excitation pulses recorded before and after the registration of the corresponding fluorescence signal. For a given curve, the two lifetimes are in close agreement, the remaining differences being mainly due to some systematic effects on both fluorescence curves and exciting pulse shapes, including the transit time jitter of the PMT (1.2 ns), the intensity non-linear response of the whole detection system and the randomicity of the stray light of the excitation laser when registering the excitation pulses. The mean value of the two lifetimes and its standard deviation were taken to be the radiative lifetime revealed by the curve and its systematic error, respectively. For each level, 15–30 curves were recorded under different experimental conditions, and the average values of the lifetimes and the systematic errors, evaluated from the curves recorded after a sufficiently long delay from the ablation pulse through the above-mentioned method, were taken as the final lifetime and systematic error results, respectively.

4 RESULTS AND DISCUSSION

The lifetimes of 53 odd-parity highly excited levels over the energy range 26 599.08–36 107.66 cm^{-1} in singly ionized samarium, which were measured in this work, are listed in Table 1. The quoted error bars consist of both the statistical uncertainties from different recordings and the systematic systematic errors. In addition, so far the electronic configurations concerning the vast numerous levels under study have not been assigned in any literature; therefore, only the total angular momentum J values of the levels are presented in Table 1. For each level in this table, the J value and the energy position were obtained from the book compiled by Martin et al. (1978).

Due to the complexity of the spectrum of Sm and the linewidth of the exciting laser (about 0.08 cm^{-1}), some odd-parity highly excited levels of Sm II could be excited at the same time from different original levels; so, the investigations of their lifetimes had to be abandoned. The measured lifetime values are in the range 10.9–271.4 ns. In this measurement, 31 lifetimes were reported for the first time, and the others are well coincident with those reported in previous papers. In these papers, most of the lifetime values of Sm II can be obtained from results reported by Lawler et al. (2006). All our measurements are within 10 per cent of theirs, and the majority are well within 5 per cent except for the 31 774.52 and 34 418.95 cm^{-1} levels. For the two levels, the lifetimes reported by Xu et al. (2003) are well coincident with ours. In addition, we also get a good agreement with the other lifetimes measured by various methods. Therefore, we believe the confidence in these investigations of lifetimes on Sm II is high. According to the statistics, the uncertainties of lifetimes are not larger than ±10 per cent.

In this work, the large J values of the investigated levels are mainly owing to the larger total orbital angular momentum L of the $4f^n$ subshell. For such complicated ionic systems, theoretical investigations are still challenging. Our experimental results on the lifetimes would be valuable for theoretical exploration and should be especially helpful for a more complete analysis and the eventual assignment of correlative levels (Lawler et al. 2006), as well as for the determination of transition probabilities in Sm II when accurate branching fractions will become available.

5 CONCLUSIONS

Radiative lifetimes of 53 odd-parity levels of Sm II, in which 31 results were reported for the first time, have been measured by the time-resolved LIF technique in laser-produced plasma. In our radiative lifetime investigation, seven upper levels above 35 000 cm^{-1}, where the knowledge is inadequate, have been included. This work would be helpful in gaining some insight into the ionic structure and radiative properties as well as for theoretical research of single ionic samarium.

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