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Measurements of radiative lifetimes, branching fractions, transition probabilities, and oscillator strengths for Eu II and Eu III levels

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ABSTRACT

Radiative lifetimes of 11 levels of Eu II in the energy range 34 923.43–46 029.14 cm^{-1} and those of six levels in the 4f^{6}5d configuration of Eu III in the range from 39 636.88 to 42 530.91 cm^{-1} were measured by time-resolved laser-induced fluorescence spectroscopy in laser-induced plasma. The obtained lifetimes range from 33 to 760 ns. To our knowledge, six levels for Eu II and three levels for Eu III are reported for the first time. Transition probabilities and oscillator strengths for 24 lines of Eu II and five lines of Eu III relative to the studied levels were derived by combining the obtained lifetimes with branching fractions measured in the emission spectrum of a hollow cathode lamp.

Key words: atomic data – atomic processes – radiative transition – instrumentation: spectrographs – methods: laboratory – techniques spectroscopic.

1 INTRODUCTION

Europium (Eu, $Z = 63$) is one of the rare earth (RE) elements and characterized by a 4f^{7}6s^2 ground configuration. Eu is composed of two isotopes of ^{151}Eu (47.8 per cent) and ^{153}Eu (52.2 per cent) and is a nearly pure *r*-process element (Mucciarelli et al. 2008). It has a complex electronic structure due to many electrons in an open 4f subshell. Its atomic energy levels have been extensively investigated (Martin, Zalubas & Hagan 1978; Wyart et al. 2008). In astrophysics, reliable oscillator strengths are important atomic radiative parameters in analysis of stellar spectra to learn chemical compositions and physical process in stars (Mashonkina, Ryabtsev & Ryabchikova 2002; Mykhailytska 2010).

Recently, a number of Eu II and Eu III spectral lines have been observed in stellar spectra (Elkin, Kurtz & Mathys 2011; Reddy, Giridhar & Lambert 2012; Ryabchikova & Romanovskaya 2017). The doubly ionized ions of RE elements are usually dominant in stellar atmospheres, but the abundance determinations were often based on the second spectra due to the lack of the reliable third spectral data (Popović, Dimitrijević & Ryabchikova 1999). In addition, Ryabchikova et al. (1999) indicated that the Eu abundance deduced from an astrophysical derived *f* value of an Eu III line largely differs from the results based on Eu II lines. Therefore more experimental oscillator strengths of Eu III lines are important to settle this kind of abundance inconsistency.

The radiative lifetimes of Eu II were measured by different methods including the multichannel method of delayed coincidences (Blagoev et al. 1978; Penkin, Gorshkov & Komarovskii 1984), the time-resolved laser-induced fluorescence (TR-LIF) method (Meyer et al. 1981; Zhang et al. 2000; Den Hartog, Wickliffe & Lawler 2002; Zhang et al. 2011b), and the laser-ion beam technique using intracavity excitation and tandem pumping (Arnesen et al. 1983). Very recently, Wang et al. (2013) measured the lifetimes of 30 high-excited levels in Eu II by TR-LIF technique and then by combining lifetime values with branching fractions, the transition probabilities and oscillator strengths for 18 transitions were deduced. They also gave a brief summary of the present research status of Eu II radiative parameters.

For Eu III, so far only three levels in the 4f^{6}5d configuration with the energies of 39 769.05, 40 870.60, and 42 084.25 cm^{-1} were measured by Zhang et al. (2000) and Den Hartog et al. (2002), respectively, using the TR-LIF technique. In experiments, it is difficult to accurately measure radiative lifetimes for doubly ionized atoms because it is hard to achieve stable ion source and avoid various detrimental effects during the lifetime measurements (Zhang et al. 2001). Therefore, theoretical calculations were usually used for intensive studies of the radiative parameters of doubly ionized atoms. In 2002, using Cowan's RCN-RCG-RCE codes (1981) and single-configuration approximation, Mashonkina et al. (2002) reported theoretical calculations of oscillator strengths for the 4f^7–(4f^{6}5d + 4f^{6}6s) transitions in Eu III. Very soon, in consideration of configuration interaction and core-polarization effects, Quinet & Biémont (2003) calculated transition probabilities and oscillator strengths of Eu III using Hatree–Fock method with semi-empirical corrections. The lifetimes of 39 769.05, 40 870.60,

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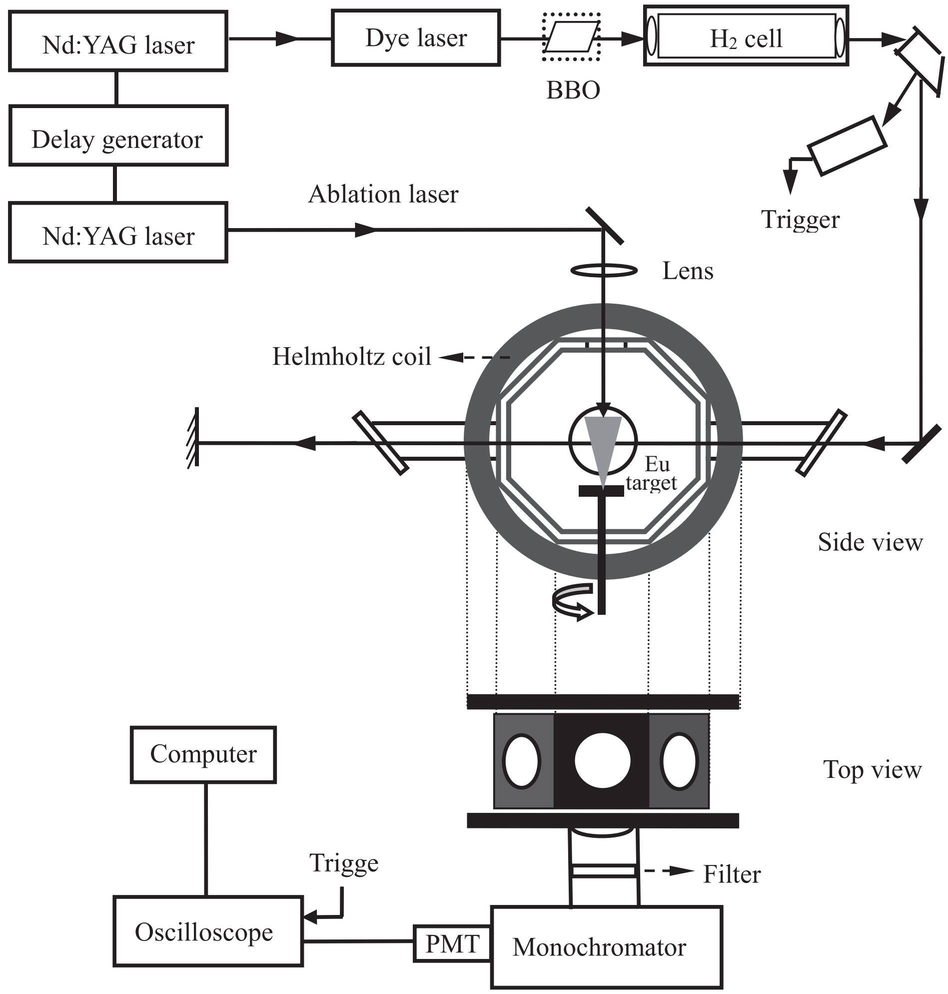

Figure 1. Experimental set-up for lifetime measurements.

Table 1. Measured lifetimes of excited levels of Eu II and comparison with previous results.

	Upper level			Lower level		Excitation laser		λ_{Obs} (nm)	Lifetime (ns)	
Config.	Term	J	E (cm^{-1})	J	E (cm^{-1})	λ_{Exc} (nm)	Non-linear scheme/Dye		This work	Previous
	106	3	34 923.43	4	0	286.341	2ω/6G	286	387(18)	400(23)[a]
	107	4	35 045.98	4	0	285.339	$2\omega + a$/DCM	409	760(70)	
	108	3	35 440.88	4	0	282.160	2ω/6G	394	35.5(2.8)	33(2)[a], 32.2(1.6)[b], 35(3)[c]
$4f^6(^7F)5d^2(^3F)$	x ^{9}P	3	36 628.00	3	1669.21	286.051	2ω/6G	380	65(5)	62.4(3.1)[b], 64(5)[c]
	z ^{7}D?	4	37 010.70	3	1669.21	282.954	2ω/6G	270	69(5)	65(3)[a], 62.2(3.1)[b]
	z ^{7}D?	3	37 167.90	3	1669.21	281.701	2ω/6G	367	86(7)	88(5)[a], 82.4(4.2)[b], 78.6(1.7)[d]
	136	4	41 666.81	3	1669.21	250.015	$2\omega + 2a$/DCM	251	680(70)	
	138	2	42 391.03	3	1669.21	245.569	$2\omega + 2a$/DCM	245	341(25)	
	y ^{9}D?	2	42 438.56	3	1669.21	245.282	$2\omega + 2a$/DCM	245	510(50)	
	y ^{9}D?	4	44 075.84	3	1669.21	235.812	$3\omega + s$/DCM	299	677(40)	
	y ^{9}D?	6	46 029.14	5	10 643.48	282.600	$2\omega + a$/DCM	343	400(50)	

Notes.
[a]From Wang et al. (2013).
[b]From Den Hartog et al. (2002).
[c]From Zhang et al. (2000).
[d]From Zhang et al. (2011b).

Table 2. Measured lifetimes of excited levels of Eu III and comparison with previous results.

Config.	Upper level Term	J	E (cm^{-1})	Lower level J	E (cm^{-1})	Excitation laser $\lambda_{Exc.}$ (nm)	Non-linear scheme/Dye	$\lambda_{Obs.}$ (nm)	Lifetime (ns) This work	Previous Exp.	Calc.
$4f^6(^7F)5d$	^8P	5/2	39 769.05	7/2	0	251.452	$2\omega + a$/6G	251	68(6)	65(7)[a], 70.8(3.5)[b]	62[c]
$4f^6(^7F)5d$	^8P	7/2	40 870.60	7/2	0	244.675	$2\omega + 2a$/DCM	245	43(4)	46(5)[a], 44.0(2.2)[b]	46[c]
$4f^6(^7F)5d$	^8P	9/2	42 084.25	7/2	0	237.619	$3\omega + s$/DCM	238	33(3)	36(4)[a], 34.7(1.7)[b]	38[c]
$4f^6(^7F)5d$	^8F	5/2	39 636.88	7/2	0	252.290	$2\omega + a$/6G	252	130(10)		
$4f^6(^7F)5d$		5/2	40 897.66	7/2	0	244.513	$2\omega + 2a$/DCM	245	66(6)		
$4f^6(^7F)5d$	^6P	7/2	42 530.91	7/2	0	235.123	$3\omega + s$/DCM	235	87(7)		

Notes.
[a]From Zhang et al. (2000).
[b]From Den Hartog et al. (2002).
[c]From Quinet & Biémont (2003).

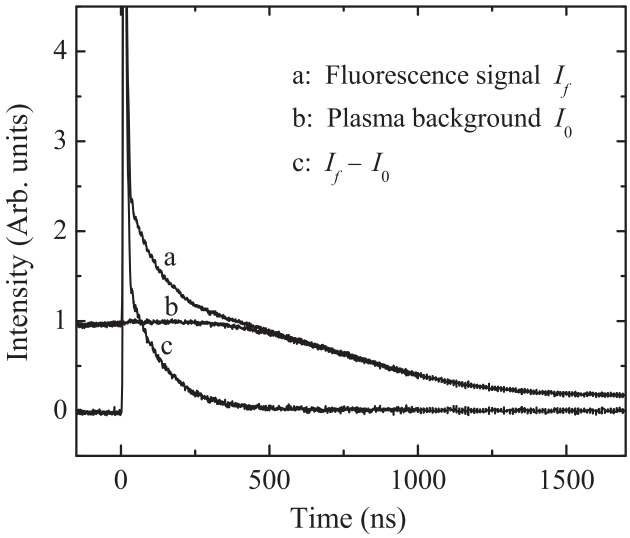

Figure 2. The fluorescence decay curve I_f of the 39 636.88 cm^{-1} level of Eu III, the plasma background I_0 and the decay curve I_0–I_f for exponential fitting.

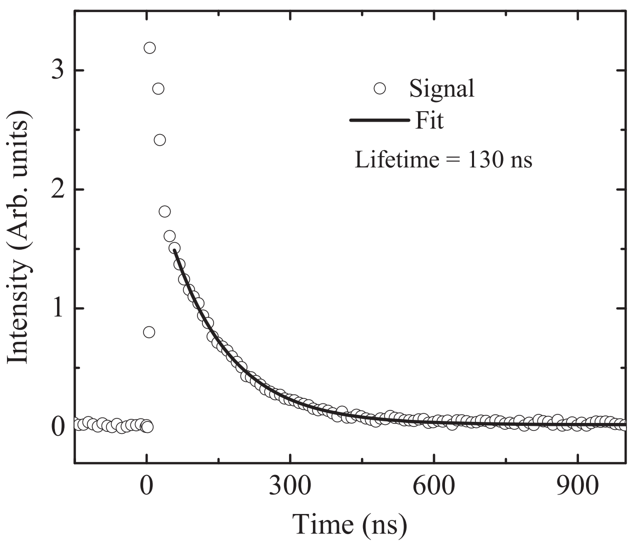

Figure 3. The decay curve I_0–I_f in Fig. 2 with an exponential fit for lifetime evaluation.

and 42 084.25 cm^{-1} levels were derived to be 65, 46, and 38 ns, respectively. Compared with the experimental values (Zhang et al. 2000), the theoretical model used by Quinet & Biémont (2003) is more reliable than Mashonkina's method (2002) for the radiative parameters of Eu III. This theoretical method was also used for other doubly ionized RE atoms, such as Er III, Dy III, Nd III (Biémont et al. 2001; Zhang et al. 2002a; Zhang et al. 2002b).

In this paper, we measure radiative lifetimes for 11 levels (34 923.43–46 029.14 cm^{-1}) in Eu II and six levels (39 636.88–42 530.91 cm^{-1}) in Eu III by the TR-LIF technique in laser-induced europium plasma. Moreover, transition probabilities and oscillator strengths for 24 transitions belonging to seven levels in Eu II and five transitions to five levels in Eu III are derived by combining the measured lifetimes and branching fractions.

2 LIFETIME MEASUREMENTS

The experimental set-up for lifetime measurements is shown in Fig. 1, which are more detailedly described in previous papers (Wang et al. 2013; Tian et al. 2016). To obtain the excitation wavelength (235.812–286.341 nm) for this work, some non-linear processes were adopted. The non-linear conversion schemes employed for different levels are shown in Tables 1 and 2. Free Eu II and Eu III ions were excited from the ground or low-lying metastable states by single-step excitation.

In this work, the delay time between the ablation and excitation lasers was adjusted by a digital delay generator (SRS DG535) and changed in a range from 5 to 15 µs for Eu II and 1.4–2.2 µs for Eu III. Among that delay time range, the fluorescence intensities changed in a relatively wide range, but the evaluated lifetimes maintained consistency within measurement uncertainties. Because of the rich spectral lines of Eu II and Eu III, great efforts have been made to avoid the co-excitation and ensure that a desired level was excited in the experiment (Tian et al. 2016). In addition, some systematic effects can induce additional errors to lifetime, so the effects (e.g. the radiation trapping, PMT non-linear response, saturation, and collision effects) were carefully considered. When the experiment conditions (e.g. the delay time, the energy of ablation laser) were appropriate together with the vacuum pressure less than 2×10^{-3} Pa, the aforesaid effects could be well eliminated. The velocities of Eu III ions are much faster than Eu II ions and Eu atoms. We took particular care to the influence of the flight-out-of-view effect for the multiply-ionized atoms, especially for the longer lifetimes. This effect was checked by changing the slit width and choosing a suitable delay time.

Table 3. Branching fractions, transition probabilities, oscillator strengths of Eu II, and comparison with previous results. The dashes denote that the transitions cannot be observed in our measurements.

Transition λ_{ki} (nm)$_{air}$	Lower level J	E_i (cm^{-1})	R_{ki} This work	R_{ki} Previous	g_kA_{ki} (10^7 s^{-1}) This work	g_kA_{ki} Previous	log(g_if_{ik}) This work	log(g_if_{ik}) Previous
				26 172.83 cm^{-1}, J = 5, τ = 6.2(3) ns[a]				
381.967	4	0	0.854(35)	0.845(34)[b], 0.836(12)[c], 0.800(11)[d]	152(10)	150(9)[b], 148.5[c], 139.7[d]	0.52(3)	0.52(3)[b], 0.51[c], 0.491(28)[e]
630.342	4	10 312.82	0.007(1)	0.0065(6)[b], 0.0091(12)[c], 0.0137(11)[d]	1.2(3)	1.2(1)[b], 1.617[c], 2.42[d]	– 1.15(9)	– 1.16(5)[b], –1.02[c], – 0.854(31)[e]
643.764	5	10 643.48	0.039(2)	0.053(2)[b], 0.043(5)[c], 0.049(4)[d]	6.9(5)	9.4(6)[b], 7.7[c], 8.58[d]	– 0.37(3)	– 0.23(3)[b], –0.32[c], – 0.276(41)[e]
664.510	6	11 128.22	0.100(6)	0.095(4)[b], 0.111(12)[c], 0.138(10)[d]	18(2)	17(1)[b], 19.69[c], 24.2[d]	0.07(4)	0.10(5)[b], 0.12[c], 0.204(27)[e]
				27 104.07 cm^{-1}, J = 3, τ = 7.2(4) ns[a],				
368.843	4	0	0.119(7)	0.110(5)[b], 0.108(5)[c], 0.112(7)[d], 0.092(9)[f]	11.6(10)	10.7(8)[b], 10.5[c], 8.89[d], 9.0(14)[f]	– 0.63(4)	– 0.66(3)[b], –0.67[c], – 0.745(24)[e], –0.73[f]
393.050	3	1669.21	0.870(36)	0.879(35)[b], 0.820(8)[c], 0.839(11)[d], 0.77(4)[f]	85(6)	85.5(6)[b], 79.8[c], 66.5[d], 75(8)[f]	0.29(3)	0.30(3)[b], 0.27[c], 0.190(25)[e], 0.24[f]
581.875	2	9923.00	0.008(1)	0.007(1)[b], 0.0115(15)[c], 0.0110(8)[d], 0.014(1)[f]	0.82(10)	0.68(1)[b], 1.12[c], 0.91[d], 1.4(2)[f]	– 1.38(5)	– 1.46(7)[b], –1.25[c], – 1.347(39)[e], –1.15[f]
587.298	3	10 081.65	0.003(1)	0.0045(6)[b], 0.0058(6)[c], 0.0065(5)[d], 0.008(1)[f]	0.27(4)	0.44(6)[b], 0.56[c], 0.511[d], 0.78(14)[f]	– 1.86(6)	– 1.65(6)[b], –1.54[c], – 1.585(30)[e], –1.40[f]
595.384	4	10 312.82	–	0.005(1)[f]	–	0.385[d], 0.47(12)[f]	–	– 1.699(65)[e], –1.60[f]
989.830	4	17 004.06	–	0.0596[f]	–	5.80[f]	–	– 0.070[f]
1003.422	3	17 140.87	–	0.0282[f]	–	2.74[f]	–	– 0.383[f]
1014.299	2	17 247.67	–	0.0209[f]	–	2.04[f]	–	– 0.503[f]
				34 923.43 cm^{-1}, J = 3, τ = 387(18) ns[g]				
286.257	4	0	0.730(33)		1.32(9)		– 1.79(3)	
300.626	3	1669.21	0.109(13)		0.20(3)		– 2.57(6)	
406.214	4	10 312.82	0.161(10)		0.29(3)		– 2.14(4)	
				35 045.98 cm^{-1}, J = 4, τ = 760(70) ns[g]				
285.255	4	0	0.007(3)		0.010(3)		– 4.0(2)	
404.202	4	10 312.82	0.881(37)		1.04(11)		– 1.59(5)	
409.678	5	10 643.48	0.112(9)		0.13(2)		– 2.48(6)	
				37 010.70 cm^{-1}, J = 4, τ = 69(5) ns[g]				
270.112	4	0	0.612(36)		8.0(8)		– 1.06(4)	
282.871	3	1669.21	0.277(12)		3.6(3)		– 1.36(4)	
371.240	3	10 081.65	0.033(4)		0.43(6)		– 2.05(6)	
374.456	4	10 312.82	0.027(9)		0.35(11)		– 2.1(2)	
379.151	5	10 643.48	0.051(9)		0.67(13)		– 1.84(9)	
				41 666.81 cm^{-1}, J = 4, τ = 680(70) ns[g]				
239.926	4	0	0.224(19)		0.30(4)		– 2.59(6)	
249.940	3	1669.21	0.255(24)		0.34(5)		– 2.50(6)	
322.245	5	10 643.48	0.521(27)		0.69(8)		– 1.97(5)	
				46 029.14 cm^{-1}, J = 6, τ = 400(50) ns[g]				
282.517	5	10 643.48	0.293(18)		0.95(13)		– 1.94(6)	
286.441	6	11 128.22	0.707(31)		2.3(2)		– 1.55(5)	

Notes.
[a]From Den Hartog (2002).
[b]From Wang et al. (2013).
[c]From Lawler et al. (2001).
[d]From Karner et al. (1982).
[e]From Biémont et al. (1982).
[f]From Zhang et al. (2000).
[g]Measured in this work.

Table 4. Branching fractions, transition probabilities, and oscillator strengths of Eu III. The dashes denote the transitions cannot be observed in our measurements. The lifetime values were measured in this work.

Upper level Config.	E_k (cm^{-1})	τ (ns)	Lower level J	E_i (cm^{-1})	Transition λ_{ki} (nm)$_{air}$	R_{ki}	g_kA_{ki} (10^7 s^{-1})	log(g_if_{ik})
4f^6(^{7}F)5d $J = 5/2$	39 769.05	68(6)	3.5	0	251.376	1	8.8(8)	−1.08(4)
			3.5	28 200.06	864.143	–		
			2.5	28 628.54	897.625	–		
4f^6(^{7}F)5d $J = 7/2$	40 870.60	43(4)	3.5	0	244.601	1	18.6(18)	−0.78(5)
			3.5	28 200.06	789.015	–		
			2.5	28 628.54	816.632	–		
4f^6(^{7}F)5d $J = 9/2$	42 084.25	33(3)	3.5	0	237.546	1	30.3(28)	−0.59(4)
			3.5	28 200.06	720.046	–		
4f^6(^{7}F)5d $J = 5/2$	39 636.88	130(10)	3.5	0	252.214	1	4.6(4)	−1.36(4)
			3.5	28 200.06	874.129	–		
4f^6(^{7}F)5d $J = 5/2$	40 897.66	66(6)	3.5	0	244.439	1	9.1(9)	−1.09(5)
			3.5	28 200.06	787.333	–		
			2.5	28 628.54	814.830	–		

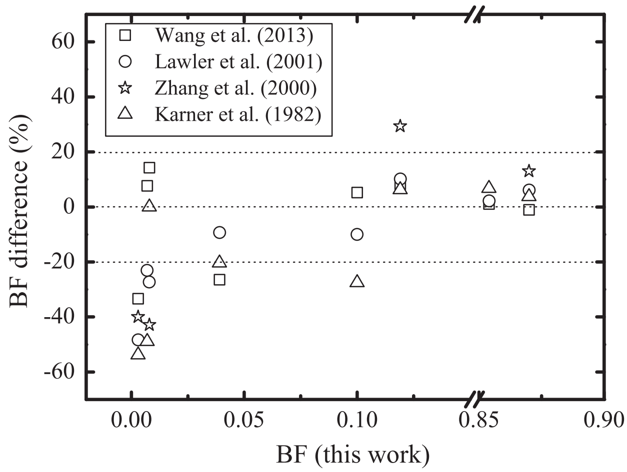

Figure 4. Comparison of our Eu II BFs to previous results in literature as a function of our BFs.

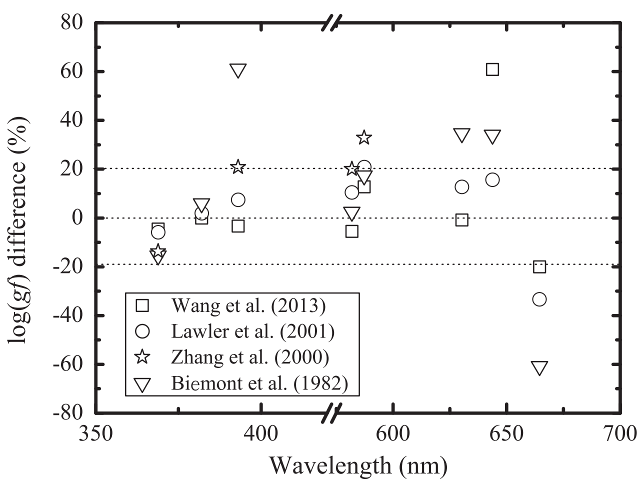

Figure 5. Comparison of our Eu II log(*gf*) values to previous results in literature as a function of wavelength. The symbols denote the differences (ours–other's)/other's.

The fluorescence of Eu III occurred at shorter delay time when the recombination between electrons and ions were dramatic, so that the signals were strongly influenced by the plasma light. This plasma light can be effectively suppressed by applying an external magnetic field (Zhang et al. 2001). Thus, up to about 200 Gauss was applied in the plasma region by a pair of Helmholtz coils. Besides, a narrow-band filter was placed in front of the monochromator to filter stray laser light and most of plasma light. The residual background light I_0 including the stray excitation laser and the plasma light is still needed to be deducted from fluorescence signal curve I_f before fitting processes, which can be seen in Fig. 2. Lifetimes of Eu III levels were evaluated from the I_f–I_0 curves by a least-squares exponential fitting. A decay curve I_f–I_0 obtained by subtraction of the background light for the 39 636.88 cm^{-1} level in Eu III with an exponential fitting are shown in Fig. 3. For each level, the average value of the lifetimes evaluated from more than eight decay curves without influence of the above effects together with the corresponding pure background (i.e. I_f–I_0 curves) recorded in different conditions was severed as the final lifetime.

3 BRANCHING FRACTION MEASUREMENTS

The branching fraction measurements process is basically the same as that used by Fan et al. (2014) and Wang et al. (2013). The emission spectra of a europium hollow cathode lamp (HCL) with argon carrier gas were collected by a grating spectrometer, equipped with three gratings having groove densities of 2400, 1800, and 600 lines mm^{-1}, respectively. This spectrometer works in a response range from 185 to 900 nm and has a spectral resolution of 0.015, 0.03, and 0.06 nm corresponding to the above gratings, respectively, when the entrance and exit slits are 10 μm. In addition, one filter was used between the lamp and the spectrometer to avoid the overlap of high-order spectra.

The theoretical bases for BF measurements was descried in detail by Jiang et al. (2012). In order to check the self-absorption effect in the HCL, the Eu–Ar emission spectra were measured at different discharge currents (6, 8, and 10 mA), and by analysing the line strength ratios, there is no self-absorption effect observed in BF results. For a level, all observed branching fractions were recorded within the shortest possible time to avoid the intensity drift in emission source and its BFs were measured for four times. Their average value was taken as the final result and the BFs' uncertainties are determined based on the method described by Fan et al. (2014).

4 RESULTS AND DISCUSSION

Radiative lifetimes of 11 levels in Eu II and six levels in Eu III were measured by TR-LIF technique. The levels and the results are listed in Tables 1 and 2, respectively, with previously reported results for comparison. In the tables, the energy positions, electronic configurations, terms, and J values are from the compilations of Martin et al. (1978). The error bar of the lifetimes contains systematical error from fitting processes and statistical uncertainty resulting from the deviations of different recordings. To our knowledge, the lifetimes of six Eu II levels and three Eu III levels are reported for the first time.

In Table 1, the uncertainties of most results are within 10 per cent, except that the 41 666.81 and 46 029.14 cm^{-1} levels in Eu II have some little larger errors 10.3 per cent and 12.5 per cent, respectively. It is seen that our results are in good agreements with previous ones by other groups (Zhang et al. 2000; Den Hartog et al. 2002; Zhang et al. 2011b; Wang et al. 2013).

In Table 2, six lifetime results for Eu III excited levels range from 33 to 130 ns and all their uncertainties are about 10 per cent. Using the laser plasma as an ionic source, it is difficult to achieve 5 per cent or higher accuracy for Eu III levels due to the influence of plasma background light. Our results are in good agreement with the previously reported data by Zhang et al. (2000), Den Hartog et al. (2002), and Quinet & Biémont (2003), and the average of root mean square differences are 7.1, 4.1, and 10 per cent, respectively, regarding our results as references.

If taking the ground states as initial states, the excitation wavelengths of the 42 084.25 cm^{-1} level of Eu III and the 42 087.02 cm^{-1} level of Eu I are 237.619 and 237.603 nm, respectively. These two levels could be easily co-excited due to very close excitation wavelengths. In the measurement, the maximum fluorescence signal was observed at 1.5 µs delay time which accorded with the feature of Eu III described in Section 2. The lifetime of 42 087.02 cm^{-1} measured by Zhang et al. (2011a) and Den Hartog et al. (2002) at delay time longer than 10 µs is 31.9 (0.6) and 30.4 (1.5) ns, respectively. Therefore, it is reasonable to think that the level we measured in this paper is 42 084.25 cm^{-1} of Eu III.

In our experiment, the BFs of 24 lines of seven Eu II levels and five lines of five Eu III levels were measured, and the corresponding transition probabilities gA and oscillator strengths log(gf) were deduced by combining the measured lifetime with BF results. The BFs, gA, and log(gf) results for Eu II and Eu III are listed in Tables 3 and 4, respectively. The uncertainties of the gA and log(gf) were evaluated by error transfer theory from the uncertainties of adopted experimental lifetimes and BFs.

Previously reported BFs, gA, and log(gf) of eight Eu II lines concerning two upper levels are listed in Table 3 for comparison. In the papers of Karner et al. (1982) and Lawler et al. (2001), the transition probabilities were unweighted (A), but the weighted values (gA) are listed in our paper. For clarity, BF comparison by the differences (ours–other's)/other's are plotted as a function of our BFs as shown in Fig. 4. It is seen that almost all the differences larger than 20 per cent belong to weak branches ($R_{ki} < 0.1$).

Fig. 5 presents the comparisons between our and previous log($g_i f_{ik}$) results (Biémont et al. 1982; Zhang et al. 2000; Lawler et al. 2001; Wang et al. 2013;). Most differences between our results and the published data are less than 20 per cent. There are a few cases of discrepancies larger than 20 per cent. First, for the 27 104.07 cm^{-1} level, the four lines with longer wavelengths cannot be observed by us, while the three infrared lines were not measured by Zhang et al. (2000) and thus they quoted the results by Komarovskii (1991). This is responsible for the differences of gA and log(gf) between Zhang et al. (2000) and this paper. Secondly, the calculated log(gf) results by Biémont et al. (1982) have larger differences from our results. Finally, for the very weak lines (BFs < 0.04), the relative differences are also rather large.

For Eu III, according to the selection rules of electric dipole transition about the J values and parities, all 13 presumptive transition channels from the five studied upper levels are presented in Table 4. In the scanning range of our spectrometer (185–900 nm), for each level only one transition branch can be observed, which are listed in the table. To our best knowledge, the experimental gA and log(gf) values of Eu III determined in this work are reported for the first time.

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