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Radiative properties of Eu II and Eu III obtained from lifetime and branching ratio measurements

Zhang Zhiguo†, Z S Li‡, H Lundberg‡, K Y Zhang†, Z W Dai†, Jiang Zhankui† and S Svanberg‡

† Department of Physics, Jilin University, Changchun 130023, People's Republic of China
‡ Department of Physics, Lund Institute of Technology, Box 118, S-211 00 Lund, Sweden

E-mail: Li.Zhongshan@fysik.lth.se

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Abstract. Radiative lifetimes of nine levels in Eu II ($4f^7 6p$, $4f^6 5d6s$ and $4f^6 5d^2$) and Eu III ($4f^6 5d$) have been measured using time-resolved laser-induced fluorescence spectroscopy on a laser-produced europium plasma. Oscillator strengths for a number of Eu II lines have been derived by combining the lifetimes with branching ratios measured in the emission spectrum of a hollow-cathode lamp.

1. Introduction

Radiative properties of atomic and ionic species are important for determining the abundance of elements in stellar atmospheres and testing theories of nuclear synthesis. In particular, data for europium ions are required in the study of the magnetic Ap-stars, which exhibit an over-abundance of europium of up to 10^4 relative to the solar concentration. Radiative lifetimes and transition probabilities are also needed for the search and selection of suitable laser schemes, for studying plasmas in thermonuclear facilities, and for testing atomic wavefunctions.

The purpose of the present investigation is to obtain absolute oscillator strengths for europium experimentally, through measurements of lifetimes and branching ratios. Previous lifetime data are limited to seven levels of Eu II studied by different methods. Meyer *et al* (1981) have measured the lifetimes of six levels of Eu II in the $4f^7 6p$ configuration by pulsed dye laser excitation and time-resolved observation of the fluorescence light in a discharge cell. The lifetimes of the same levels were also measured by Arnesen *et al* (1983) with an intra-cavity excitation beam–laser technique. Using the method of electron impact excitation and delayed-coincidence detection, Penkin *et al* (1984) measured the lifetimes of the same six levels and of one level of the $4f^6 5d6s$ configuration of Eu II. No lifetime measurements for Eu III have been reported in literature up to now. Information on the oscillator strengths and electronic transition probabilities for some Eu II transitions have been given by Klimkin *et al* (1978), Biémont *et al* (1982), Migdalek *et al* (1984), and reviewed by Komarovskii (1991).

In this paper, we report experimental lifetime values of six levels in Eu II and three levels in Eu III, which were measured in the UV/VUV laboratory of the Lund Laser Center (Svanberg *et al* 1994). Branching ratios for the six Eu II levels were measured at Jilin University. The oscillator strengths have been deduced by the combination of the lifetime values and the branching ratio data. Most of these atomic data are, to our knowledge, reported for the first time.

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Table 1. Radiative lifetime measurement of Eu II and Eu III levels.

Level[a]	Energy (cm^{-1})	Excitation laser		Fluorescence	Lifetime (ns)	
		λ_{vac} (nm)	Nonlinear scheme	λ_{vac} (nm)	This work	Previous
Eu II						
$4f^7 6p\ (\frac{7}{2}, \frac{3}{2})_3$	27 104.07	368.98	2ω + S	393.2	7.1(4)	7.4(2)[b] 8.8(5)[c] 8.5(9)[d]
$4f^6 5d6s\ y\ ^9P_3$	34 393.57	305.58	2ω	290.8	21(2)	
$4f^6 5d6s\ y\ ^9P_4$	35 527.02	281.48	2ω + AS	281.5	14.5(9)	14.7(7)[d]
$4f^6 5d6s\ y\ ^9P_5$	36 648.95	272.86	2ω + AS	272.9	14.8(9)	
$J = 3$	35 440.88	282.16	2ω + AS	282.2	35(3)	
$4f^6 5d^2\ x\ ^9P_3$	36 628.00	273.06	2ω + AS	286.0	64(5)	
Eu III						
$4f^6 5d\ ^8P_{5/2}$	39 769.05	251.45	2ω + 2AS	251.5	65(7)	
$4f^6 5d\ ^8P_{7/2}$	40 870.60	244.67	2ω + 2AS	244.7	46(5)	
$4f^6 5d\ ^8P_{9/2}$	42 084.25	237.62	3ω + S	237.6	36(4)	

[a] Martin *et al* (1978).
[b] Arnesen *et al* (1983) (beam–laser measurements).
[c] Meyer *et al* (1981) (time-resolved LIF).
[d] Penkin *et al* (1984) (delayed-coincidence technique).

2. Lifetime measurements

Time-resolved laser-induced fluorescence spectroscopy was employed for the present lifetime measurements of levels in Eu II and Eu III. Selective excitation was realized by pumping the desired upper levels of the europium ions with a tunable UV laser pulse. By means of an exponential fitting to the time-resolved fluorescence, which was recorded in selected decay channels by a fast-detection system, reliable lifetime values were directly obtained.

In order to obtain narrow-bandwidth, short-duration laser pulses, a stimulated Brillouin scattering (SBS) compressor was utilized (see Li *et al* 1999a for details). The second harmonic of 8 ns radiation bursts from a seeder-injected Nd:YAG laser (Continuum NY-82) were compressed before being sent to pump the dye laser (Continuum Nd-60) operating with DCM dye. The output from the dye laser, tunable from 615 to 660 nm, had a pulse energy of 10–20 mJ and a pulse duration of about 1 ns. Different kinds of nonlinear processes were used to obtain radiation on wavelengths needed for the present experiment (230–370 nm). The second harmonic (2ω) was produced by frequency doubling in KDP, the third harmonic (3ω) was produced by frequency mixing of the second harmonic with the fundamental frequency in BBO. The first and second Raman shifted anti-Stokes or Stokes components (AS, 2AS, S) were generated in a H_2 gas cell. The schemes utilized for different levels are indicated in table 1. The excitation was performed from the ground level or a low-lying metastable level.

The laser-produced plasma technique (see, e.g., Li *et al* 1999b) was employed to prepare perturbation-free Eu^+ and Eu^{2+} ions. The 532 nm laser pulses provided by a separate Nd:YAG laser (Continuum Surelite) were focused perpendicularly onto a rotating metallic europium target in the vacuum chamber to produce the plasma. About 10 mm above the ablation spot, the excitation laser beam was sent through the plasma. Laser-induced fluorescence from the selected upper levels were detected by a Hamamatsu 1564U micro-channel-plate (MCP) photomultiplier tube (200 ps rise time) after wavelength selection in a spectrometer. To make sure that the right level was excited, all the possible decay channels were checked by tuning the

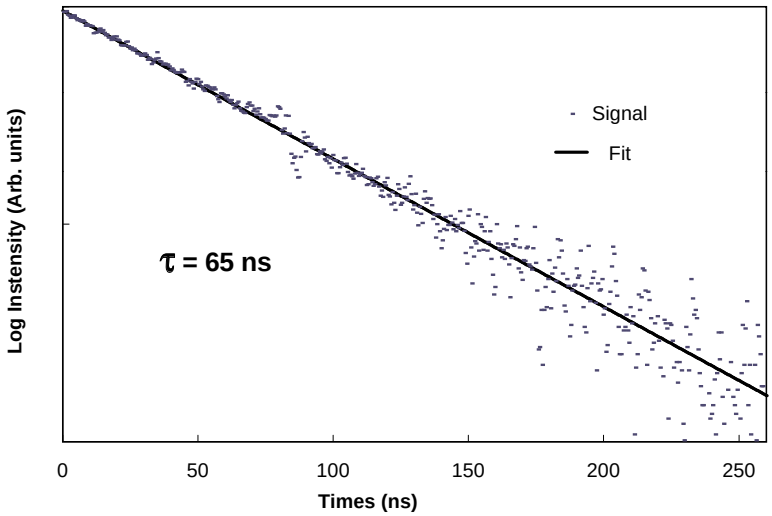

Figure 1. The logarithm of the detected fluorescence signal from the $4f^6 5d\ ^8P_{5/2}$ state with an exponential fit which gives a lifetime of 65 ns.

monochromator, and then proper transitions were selected for recording the decay curves (see table 1). A digital transient recorder (Tektronix Model DSA 602) was used to perform the data acquisition. To ensure a linear response of the detection system, only sufficiently weak signals were detected from each pulse and an average of 4000 pulses was performed to achieve the necessary signal-to-noise ratio.

By changing the ablation pulse energy, the size of the focus point, the distance above the target surface and the delay time between ablation and excitation pulses, the plasma density and temperature in the observed point could be effectively adjusted. Measurements under different plasma conditions were performed to avoid systematic errors caused by collisional quenching and radiation trapping. A strong magnetic field was also utilized to check the effects from Zeeman quantum beats. Possible flight-out-of-view effects were specially investigated by changing the slit width and position of the monochromator. A pure exponential fit was performed to obtain the lifetime values. Around 20 curves for each level have been recorded for near perturbation-free conditions and the averaged lifetime value was adopted as the final result. The logarithmic plot of a decay curve for the Eu III $4f^6 5d\ ^8P_{5/2}$ state is shown in figure 1. The error bars given in table 1 more reflect a conservative view of possible residual systematic effects than the statistical spread of the data.

3. Branching ratio measurements

Branching ratios were measured from spectra recorded with a double grating monochromator, which has a spectral resolution of 0.001 nm and a scanning range of 200–900 nm. The source of the spectra was a hollow-cathode discharge lamp with Eu powder in the cathode cavity. The Eu III spectrum are not observed with the present source. A computer controlled the scanning of the monochromator and the spectral data were stored in a computer file. A part of the spectrum is shown in figure 2. As a test of self-absorption in the source, three spectra were recorded at different discharge currents, and all branching ratios were measured from each of the three spectra. No significant re-absorption effects were observed for the Eu II transitions studied.

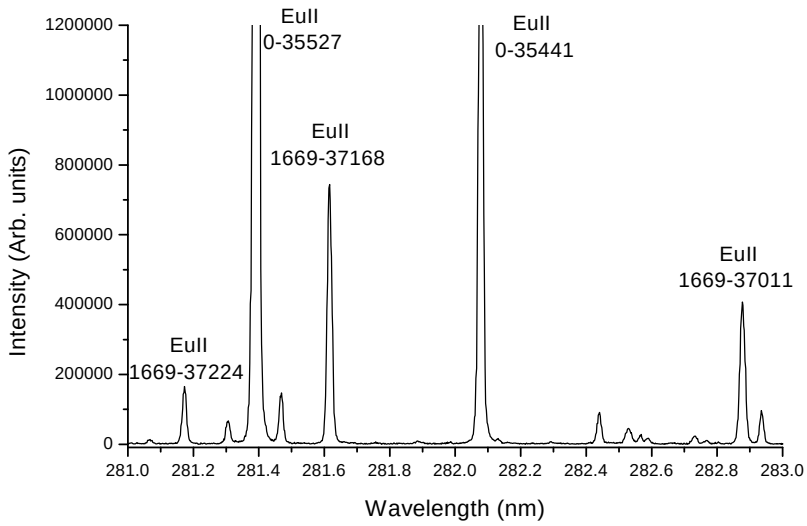

Figure 2. Part of the Eu II spectrum of a hollow-cathode discharge lamp.

The areas under the line profiles were integrated with a computer code to obtain the observed line intensity $A(\lambda)$. The relative line intensity was then given by $I(\lambda) = A(\lambda)/E(\lambda)$, where $E(\lambda)$ is the spectral response of the analysis and detection system, which was measured using a standard lamp calibrated in the Changchun Institute of Optics and Fine Mechanics in China. The branching ratio $R_{ki}(\lambda_{ki})$ for the transition λ_{ki} is then

$$R_{ki}(\lambda_{ki}) = \frac{\lambda_{ki} I(\lambda_{ki})}{\sum_i \lambda_{ki} I(\lambda_{ki})},$$

where the sum in the denominator includes all transitions from the same upper level. The data of the Eu II energy level positions given by Martin *et al* (1978) have been used to calculate the wavelengths of electric dipole allowed transitions to identify lines in our recorded spectra.

The energy level structure in Eu II is very rich. The lowest configurations, $4f^7 6s$ and $4f^7 5d$, give rise to hundreds of levels and only the position of those where the electrons in the half-filled f-shell couple to the parent ^8S are known. These 12 levels are all below 18 000 cm^{-1} and constitute the lower levels for the transitions studied in this work. In Eu III the $4f^7\ ^6P$ and ^6I levels have been classified and have energies around 30 000 cm^{-1}. This suggests that Eu II levels belonging to subconfigurations $4f^7(^6L)6s$ (L = P, D, F, G, H and I) could be located at similar energies. Unknown infrared transitions from the upper levels in our investigation might then occur. In LS coupling these transitions are forbidden. Their contribution to the total decay can, with the present knowledge of the Eu II spectrum, hardly be calculated and their possible influence has been neglected in this work.

Due to the signal-to-noise ratio of the recorded spectra, branching ratios smaller than 0.5% were not detectable in our system. The intensities of most Eu II lines were sufficiently high so that the photon shot noise of the measurement was very small by comparison, and its effect on the final uncertainties in the branching ratio was negligible. The main contribution to the uncertainty of the measured branching ratio came from two effects. One was the long-time intensity drift of the lamp, which caused less than 1% uncertainty; the other was the intensity calibration for the spectral response of the detection system, which caused less than 5% uncertainty for the strong spectral region and less than 10% for the weak spectral region.

Table 2. Branching ratios, transition probabilities and oscillator strengths of Eu II.

E_k (cm^{-1})	E_i (cm^{-1})	λ_{ki} $(nm)_{air}$	R_{ki}	$g_k A_{ki}$ (10^8 s^{-1})	$\log(gf)$	$g_k A^a_{ki}$ (10^8 s^{-1})
27 104.07	0.00	368.844	0.092(9)	0.90(14)	−0.73	0.910
	1 669.21	393.050	0.77(4)	7.5(8)	0.24	6.65
	9 923.00	581.875	0.014(1)	0.14(2)	−1.15	0.091
	10 081.65	587.298	0.008(1)	0.078(14)	−1.40	0.051
	10 312.82	595.384	0.005(1)	0.047(12)	−1.60	0.039
	17 004.06	989.830	0.0596[a]	0.580	−0.070	0.518
	17 140.87	1003.422	0.0282[a]	0.274	−0.383	0.245
	17 247.67	1014.299	0.0209[a]	0.204	−0.503	0.182
34 393.57	0.00	290.667	0.86(5)	2.9(4)	−0.44	
	1 669.21	305.494	0.056(6)	0.19(4)	−1.58	
	9 923.00	408.539	0.035(2)	0.12(2)	−1.53	
	10 081.65	411.205	0.031(2)	0.104(15)	−1.58	
	10 312.82	415.152	0.012(2)	0.043(11)	−1.96	
35 527.05	0.00	281.393	0.79(4)	4.90(55)	−0.24	5.31
	1 669.21	295.267	0.027(3)	0.17(3)	−1.66	0.324
	10 081.65	392.888	0.013(1)	0.08(1)	−1.72	0.077
	10 312.82	396.490	0.12(1)	0.75(11)	−0.75	0.171
	10 643.48	401.759	0.038(2)	0.23(3)	−1.25	
	16 860.72	535.576	0.012(1)	0.075(11)	−1.49	
36 648.95	0.00	272.778	0.95(5)	7.1(8)	−0.10	
	10 312.82	379.599	0.011(2)	0.08(2)	−1.75	
	10 643.48	384.426	0.038(3)	0.28(4)	−1.21	
35 440.88	0.00	282.077	0.84(5)	1.7(2)	−0.70	
	1 669.21	296.020	0.088(9)	0.18(3)	−1.64	
	9 923.00	391.772	0.038(4)	0.075(12)	−1.76	
	10 081.65	394.223	0.038(4)	0.076(12)	−1.75	
36 628.00	0.00	272.934	0.50(3)	0.54(7)	−1.22	
	1 669.21	285.967	0.111(6)	0.121(15)	−1.83	
	9 923.00	374.367	0.102(6)	0.112(15)	−1.63	
	10 081.65	376.593	0.140(7)	0.15(2)	−1.49	
	10 312.82	379.901	0.151(8)	0.17(2)	−1.44	

[a] Komarovskii *et al* 1991.

Infrared transitions from the 27 104.07 cm^{-1} level were not observed in our measurements. The branching ratios for these transitions were taken from Komarovskii (1991).

The oscillator strengths were deduced using the well known formulae

$$A_{ki} = \frac{R_{ki}}{\tau_k}$$

$$g_i f_{ik} = \frac{1}{0.667\,02\sigma_{ki}^2} g_k A_{ki},$$

where $g_i f_{ik}$ denotes the absorption oscillator strength, A_{ki} the transition probability, g_k, g_i the statistical weights of the upper and lower levels and σ_{ki} the wavenumber (cm^{-1}).

The oscillator strengths are listed in table 2. The uncertainties assigned to the $g_i A_{ki}$ values were evaluated from the uncertainties in the measured lifetimes and branching ratios.

4. Discussion

The lifetimes of the levels 27 104.07 cm^{-1} and 35 527.05 cm^{-1} of Eu II are compared with previous results in table 1. Our lifetime value of the 27 104.07 cm^{-1} level is in agreement with the one obtained by Arnesen *et al*, and shorter than the values given by Penkin *et al* and Meyer *et al*. Discrepancies might be related to problems with unselective excitation and radiation trapping, respectively.

The gA values of the transitions from the levels 27 104.07 cm^{-1} and 35 527.05 cm^{-1} are compared with previous results in table 2. As can be seen, our gA values differ by about 10% from previous results. The discrepancy for the 27 104.07 cm^{-1} level mainly comes from the lifetime value used for normalization. The one, which was used by Komarovskii, is an average of the values in table 1 (see footnotes b–d). The discrepancy for the 35 527.05 cm^{-1} levels might be that in the earlier work branching ratios were taken from Corliss and Bozman (1962), a source that has a large uncertainty.

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