Eur. Phys. J. D **13**, 301–304 (2001)

**THE EUROPEAN PHYSICAL JOURNAL D**

EDP Sciences


# Lifetime measurements in Gd II and Gd III using time-resolved laser spectroscopy

Z.G. Zhang[1,a], A. Persson[1], Z.S. Li[1], S. Svanberg[1,b], and Jiang Zhankui[2]

[1] Department of Physics, Lund Institute of Technology, P.O. Box 118, 211 00 Lund, Sweden
[2] Department of Physics, Jilin University, Changchun 130023, P.R. China

Received 17 July 2000 and Received in final form 13 October 2000

**Abstract.** Natural radiative lifetimes of 20 levels (energy range between 29 000–35 000 $cm^{-1}$) in Gd II and 5 levels (energy range between 43 000–49 000 $cm^{-1}$) in Gd III have been measured using time-resolved laser-induced fluorescence in a laser-produced plasma.

**PACS.** 32.70.Cs Oscillator strengths, lifetimes, transition moments – 42.62.Fi Laser spectroscopy

## 1 Introduction

The interest of natural radiative constants of rare earth atoms and ions is mainly due to their importance in astrophysics. Most singly and doubly ionized rare earth atoms (including Gd II and Gd III) have been identified in the spectra of chemically peculiar stars [1–3]. Doubly ionized rare earth elements are particularly abundant in hot chemically peculiar stars. These circumstances have motivated some theoretical and experimental work regarding the determination of natural radiative constants of rare earth ions [4].

The element gadolinium has the most complex electronic structure among the rare earth elements due to a half-full $4f$ electron shell and the presence of a $5d$ electron. Presently, accurate atomic radiative parameters can hardly be theoretically calculated and the experimental data constitute the only source for obtaining these parameters. First results on natural radiative lifetimes for the element appeared around 1980. 16 lifetimes in Gd I were measured using the delayed-coincidence method with laser excitation [5]. In 1983 and 1986, Gorshkov *et al.* reported lifetime measurement results on 16 lifetimes in Gd I and 6 lifetimes in Gd II using the delayed-coincidence technique in intersecting beams of atoms and electrons [6,7]. Bergström *et al.* published 3 lifetimes in Gd II measured using time-resolved laser-induced fluorescence technique and gave a revised solar abundance for gadolinium [8].

To meet the needs in astrophysics, lifetime measurements for doubly-ionized rare earth elements were carried out at the Lund Laser Centre, in order to obtain abundance values of those elements in the Sun and in other stars [9–11]. The results were in good agreement with the results from theoretical calculations using the Cowan programs [12].

In the present paper, lifetime measurements for 20 levels in Gd II and 5 levels in Gd III using the time-resolved laser-induced fluorescence method are reported. In the lifetime measurements, the free ions were obtained in a laser-produced plasma. A narrow-band and short-pulse (1 ns pulse duration) dye laser followed by subsequent doubling, tripling or Stimulated Stokes Raman Scattering (SSRS) was used as a selective excitation source, and a fast detection system including a micro-channel plate photomultiplier tube and a 1 GHz bandwidth digital transient recorder was used to record the decay curves.

## 2 Experimental set-up

The experimental set-up used in the lifetime measurements is shown in Figure 1. Here only a brief description is given, because it has been described in detail elsewhere [13].

In order to measure the lifetimes of the selected levels in Gd II and Gd III, a laser operating in the wavelength ranges of 204–233 nm and 305–390 nm is required. The radiation was produced using different nonlinear schemes: frequency doubling, frequency tripling and SSRS.

532 nm laser pulses from an injection seeded Nd:YAG laser (Continuum NY-82) were passed through a Stimulated Brillouin Scattering (SBS) system. The output from the SBS system consists of 1 ns pulses with 150 mJ single pulse energy, and was used to pump a dye laser. DCM or R610+R640 dyes were used in the experiments. The dye laser output with a maximum of 20 mJ single pulse energy was frequency-doubled in a KDP crystal or frequency-tripled in a non-linear optical system consisting of a KDP

[a] e-mail: zhang.zhiguo@fysik.lth.se
[b] e-mail: sune.svanberg@fysik.lth.se

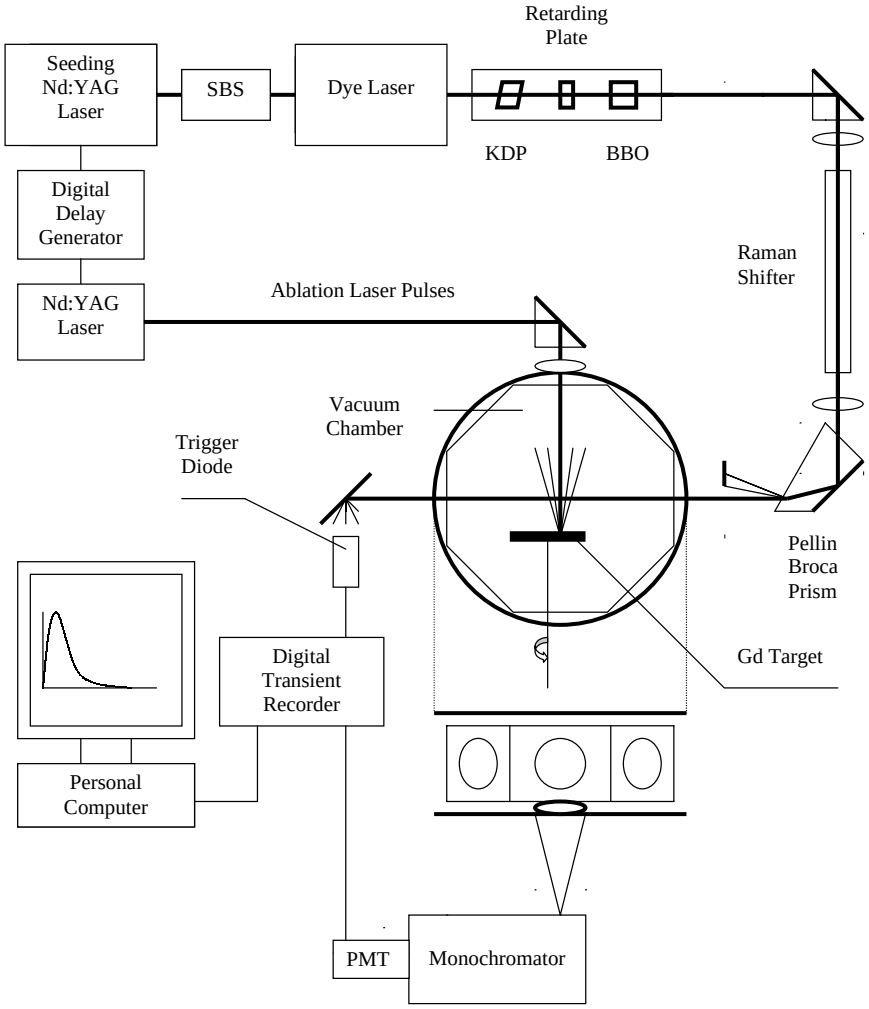


**Fig. 1.** Experimental set-up.

crystal, a retarding plate, and a BBO crystal. The first order SSRS was obtained when the frequency-doubled or frequency-tripled laser beam was focused into a hydrogen cell with 10 bar pressure. Three lifetimes in Gd III were measured with the frequency-tripled laser. The other lifetimes in Gd III were measured with the first-order SSRS from the frequency-tripled laser. Three lifetimes in Gd II were measured with the first-order SSRS using the frequency-doubled laser. The other lifetimes in Gd II were measured with the frequency-doubled system (see Tabs. 1 and 2).

In order to obtain free Gd II and Gd III ions, laser ablation using a further 532 nm pulsed laser was employed. The laser beam was focused perpendicularly onto the surface of a Gd foil, which was rotated in a vacuum chamber by a AC motor. An expanding plasma was produced above the surface of the sample. The two Nd:YAG lasers employed in the experiments were triggered externally by a digital delay generator (Stanford Research System Model 535), with the possibility to adjust the delay time between the ablation and excitation pulses. In this way, the plasma conditions can be varied.

The laser beam used to excite the ions was passed horizontally through the plasma at a distance of 5–20 mm above the sample surface. Fluorescence induced by the laser was collected by a fused-silica lens and focused to the entrance slit of a 1/8 m monochromator (resolution 6.4 nm/mm). The fluorescence from the measured level was selected by the monochromator, and was detected by a Hamamatsu 1564U micro-channel-plate (MCP) photo multiplier with a 200 ps risetime. Then the signal was acquired and averaged by a digital transient recorder (Tektronix Model DSA 602). Finally, the averaged time-resolved fluorescence decay curve was sent to a personal computer for lifetime evaluation.

## 3 Measurements and results

In order to make sure that the correct level was excited, all the decay channels were observed by tuning the wavelength of the monochromator, and the signal intensity in each channel was compared with the line intensity from the NIST Atomic Spectra Database (`http://www.physics.nist.gov/cgi-bin/AtData/lines_form`).

**Table 1.** Levels measured in Gd II with excitation scheme and results.

| energy level** designation | energy ($cm^{-1}$) | lower level ($cm^{-1}$) | $\lambda_{exc}$ $(nm)_{air}$ | $\lambda_{obs}$ $(nm)_{air}$ | $\lambda_{exc}$ laser conversion scheme | $\tau$ (ns) this work | $\tau$ (ns) previous' |
|---|---|---|---|---|---|---|---|
| $4f^7(^8S^0)5d(^9D^0)6p\ ^{10}D_{9/2}$ | 29 045.29 | 1158.9 | 358.50 | 413.04 | $2\omega + S$ | 4.8(3) | 14(1)[a] |
| | | | | 413.23 | | | |
| $4f^7(^8S^0)5d(^9D^0)6p_{9/2}$ | 29 242.25 | 633.27 | 349.44 | 409.89 | $2\omega + S$ | 7.1(3) | 6.3[b] |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^8D_{3/2}$ | 29 877.93 | 0.00 | 334.60 | 373.09 | $2\omega$ | 4.7(3) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^8D_{5/2}$ | 29 965.75 | 0.00 | 333.62 | 368.78 | $2\omega$ | 5.0(3) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^8D_{7/2}$ | 30 008.89 | 0.00 | 333.14 | 371.27 | $2\omega$ | 5.0(3) | 3.5[b] |
| $4f^7(^8S^0)5d(^9D^0)6p_{11/2}$ | 30 101.37 | 1935.3 | 354.94 | 413.04 | $2\omega + S$ | 4.3(3) | 4.3[b], 6.3(6)[c] |
| $4f^7(^8S^0)5d(^9D^0)6p_{11/2}$ | 30 366.82 | 633.27 | 336.22 | 391.65 | $2\omega$ | 4.5(3) | 6.1(6)[c] |
| $4f^7(^8S^0)6s6p(^3P^0)\ ^8P_{9/2}$ | 30 849.65 | 261.84 | 326.83 | 364.56 | $2\omega$ | 14.6(5) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^{10}D_{13/2}$ | 30 996.85 | 1158.9 | 335.05 | 409.86 | $2\omega$ | 3.9(3) | |
| $4f^7(^8S^0)6s6p(^3P^0)\ ^6P_{7/2}$ | 31 908.12 | 0.00 | 313.31 | 313.31 | $2\omega$ | 11.9(5) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^8F_{5/2}$ | 32 150.14 | 261.84 | 313.50 | 343.92 | $2\omega$ | 5.1(3) | |
| $4f^7(^8S^0)5d(^9D^0)6p_{5/2}$ | 32 260.12 | 0.00 | 309.89 | 346.73 | $2\omega$ | 5.5(3) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^8F_{7/2}$ | 32 262.79 | 0.00 | 309.87 | 342.59 | $2\omega$ | 5.4(3) | |
| $4f^7(^8S^0)5d(^9D^0)6p_{9/2}$ | 32 304.41 | 261.84 | 312.00 | 315.65 | $2\omega$ | 4.8(3) | |
| $4f^7(^8S^0)5d(^9D^0)6p_{7/2}$ | 32 490.51 | 261.84 | 310.19 | 343.98 | $2\omega$ | 4.8(3) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^8F_{11/2}$ | 32 946.20 | 1158.9 | 314.50 | 355.71 | $2\omega$ | 4.7(3) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^{10}P_{7/2}$ | 33 211.48 | 0.00 | 301.01 | 301.01 | $2\omega^*$ | 2.1(2) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^8F_{13/2}$ | 33 557.95 | 1935.3 | 316.14 | 348.13 | $2\omega$ | 4.7(3) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^{10}P_{9/2}$ | 33 596.03 | 261.84 | 299.91 | 308.20 | $2\omega^*$ | 2.5(2) | |
| $4f^7(^8S^0)5d(^9D^0)6p\ ^{10}P_{11/2}$ | 34 178.78 | 633.27 | 298.02 | 310.05 | $2\omega^*$ | 2.5(2) | |

Energy level** was downloaded from the NIST Atomic Spectra Database (`http://www.physics.nist.gov/cgi-bin/AtData/levels_form`), $\omega$ is a frequency of a DCM dye laser, $\omega^*$ is a frequency of a R610+R640 dye laser, [a] from [5], [b] from [7], [c] from [6].

**Table 2.** Levels measured in Gd III with excitation scheme and results.

| energy level** designation | energy ($cm^{-1}$) | lower level ($cm^{-1}$) | $\lambda_{exc}$ $(nm)_{air}$ | $\lambda_{obs}$ $(nm)_{air}$ | $\lambda_{exc}$ laser conversion scheme | $\tau_{thiswork}$ (ns) |
|---|---|---|---|---|---|---|
| $4f^7(^8S^0_{7/2})6p_{1/2}(7/2,1/2)_3$ | 43 019.99 | 0.00 | 232.378 | 295.554 | $3\omega + S$ | 1.9(2) |
| $4f^7(^8S^0_{7/2})6p_{1/2}(7/2,1/2)_4$ | 43 611.69 | 694.370 | 232.935 | 290.472 | $3\omega + S$ | 1.9(2) |
| $4f^7(^8S^0_{7/2})6p_{3/2}(7/2,3/2)_5$ | 47 233.93 | 694.370 | 214.803 | 262.811 | $3\omega$ | 1.4(2) |
| $4f^7(^8S^0_{7/2})6p_{3/2}(7/2,3/2)_3$ | 48 859.62 | 0.00 | 204.602 | 258.821 | $3\omega$ | 1.5(2) |
| $4f^7(^8S^0_{7/2})6p_{3/2}(7/2,3/2)_4$ | 48 339.14 | 279.320 | 208.007 | 255.390 | $3\omega$ | 1.5(2) |
| | | | | 256.446 | | |

In these measurements, the signal was also observed at different delay times in order to ascertain which ionization stage a line belonged to: neutral atoms, singly or doubly-ionized atoms. Fluorescence decay curves for lifetime determinations were usually measured in the strongest fluorescence signals.

In the measurements, attentions to different effects, which could influence lifetime measurements, were paid.

The lifetimes measured are relatively short and the frequency of possible quantum beats caused by the Zeeman effect in the earth magnetic field is too low to reasonably affect the lifetime measurements. Still, in order to check the influence of possible quantum beat oscillations, decay curves were studied under varied magnetic fields. No lifetime changes were observed.

The magic-angle method was employed in the lifetime measurements in order to eliminate the possible distortion of the fluorescence decay curve caused by alignment effects [14,15]. No effect was observed, when the polarization direction of the exciting laser was changed. In order to investigate possible flight-out-of-view effects, different slit widths of the monochromator and delay times (1.0–4 $\mu$s for Gd II and 1.0–1.5 $\mu$s for Gd III) were selected, but no effect was observed due to the short lifetimes.

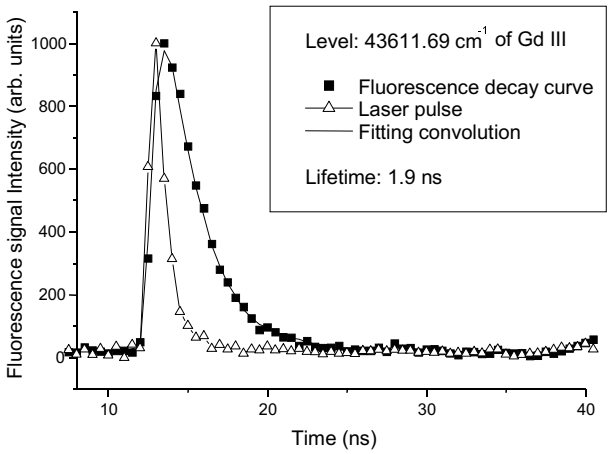


**Fig. 2.** A typical fluorescence decay curve and a convolutional fitting. The excitation pulse shape is included.

In order to avoid collision effects, saturation effects and radiation-trapping effects, the lifetime measurements were carried out under different intensities of laser excitation and different number densities of the ions. The intensity of the laser was adjusted by inserting neutral density filters in the exciting laser light path, and the number density of the ions was changed by varying the delay time. However, in the range of delay times listed above, the evaluated lifetimes of the decay curves for a level studied scattered randomly around a certain value. No obvious systematic effects were found.

In the measurements, the magnitude of the fluorescence decay signal was always kept small enough through weakening the laser intensity and reducing the number of the ions in order to avoid errors due to a nonlinear response of the detector. A decay curve needed averaging over 1000 shots in order to obtain a good signal-to-noise ratio. The temporal shape of the excitation pulse was recorded with the same detection system by observing the directly scattered light of the laser pulse from a metal rod, which was inserted into the interaction volume. The evaluated lifetime of a curve was obtained through fitting the fluorescence decay signal curve with a convolution between the detected laser pulse and an exponential function with adjustable parameters. The effects of the finite duration of the excitation laser pulse and the limited response time of the detection system were taken into account in the fitting process. Fluctuations in intensity and also in pulse length can be substantial for the excitation pulses. However, these aspects are also handled by the data taking and convolution processes, which are governed by linear mathematics. A typical decay curve and a fitted curve are shown in Figure 2. The final lifetime value for a level was given through averaging the evaluated lifetimes from about 10 curves measured at different delay times and for different laser intensities. The lifetimes of Gd II and Gd III levels and estimated error bars are listed in Tables 1 and 2. The error bars for the lifetimes of the levels reflect a statistical scattering of the evaluated lifetimes and an estimate of possible remaining systematic errors.

## 4 Discussion

Lifetimes of 20 levels for Gd II are given and compared with previous results in Table 1. The lifetime values obtained in the measurements are in very good agreement with the results measured by Bergström *et al.* [8], except for the lifetime of the level 30 008.89 $cm^{-1}$ which is slightly larger than that previously measured. However, our lifetime values are much smaller than those published by Gorshkov *et al.* [6,7], especially the lifetime of the level 29 045.29 $cm^{-1}$, which would be expected to be close to the one of the level 30 366.82 $cm^{-1}$, belonging to the same multiplet. The disagreement between the two results might be caused by unselective excitation in the earlier experiments.

This work was financially supported by the Swedish Natural Science Research Council and by the National Natural Science Foundation of China.

## References

1. S.J. Adelman, Ap. J. Suppl. **27**, 183 (1974).
2. S.J. Adelman, W.P. Bidelman, D.M. Pyper, Ap. J. Suppl. **40**, 371 (1979).
3. C.R. Cowley, G. Mathys, Astron. Astrophys. **339**, 165 (1998)
4. K.B. Blagoev, V.A. Komarovskii, At. Data Nucl. Data Table **56**, 1 (1994).
5. J. Marek, H.J. Stahnke, Z. Phys. A **298**, 81 (1980).
6. V.N. Gorshkov, V.A. Komarovskii, A.L. Osherovich, N.P. Penkin, Opt. Spectrosc. (USSR) **54**, 122 (1983).
7. V.N. Gorshkov, V.A. Komarovskii, Sov. Astron. **30**, 333 (1986).
8. H. Bergström, E. Biemont, H. Lundberg, A. Persson, Astron. Astrophys. **192**, 335 (1988).
9. Z.S. Li, Z.G. Zhang, V. Lokhnygin, S. Svanberg, T. Bastin, H.P. Garnir, P. Palmeri, E. Biémont, P. Quinet, J. Phys. B: At. Mol. Opt. Phys. (to appear).
10. Z.S. Li, J. Zhankui, Phys. Scripta **60**, 414 (1999).
11. Z.G. Zhang, Z.S. Li, H. Lundberg, K.Y. Zhang, Z.W. Dai, Z.K. Zhang, S. Svanberg, J. Phys. B: At. Mol. Opt. Phys. **33**, 521 (2000).
12. R.D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California Press, Berkeley, 1981).
13. Z.S. Li, J. Norin, A. Persson, C.-G. Wahlström, S. Svanberg, P.S. Doidge, E. Biémont, Phys. Rev. A **60**, 198 (1999).
14. P. Hannaford, R.M. Lowe, Opt. Eng. **22**, 532 (1983).
15. W. Schade, L. Wolejko, V. Helbig, Phys. Rev. A **47**, 2099 (1993).