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Radiative lifetime measurements of some Gd I levels by time-resolved laser spectroscopy

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ABSTRACT

Natural radiative lifetimes for 27 excited levels of Gd I in the energy range from 28215.140 to 43963.900 cm^{-1} were measured using time-resolved laser-induced fluorescence (TR-LIF) technique in an atom beam produced by laser-induced plasma. All the lifetimes obtained in this paper range from 8.4 to 833 ns with the uncertainties within ten percent. A comparison with a few previously reported values was performed and good agreement between them was achieved. To our best knowledge, 18 lifetimes of Gd I are reported for the first time.

1. Introduction

Atomic transition probabilities or oscillator strengths, as fundamental radiative constants of atomic or ionic species, play an important role in many fields. In astrophysics, they are required for determining accurate elemental abundances, studying Galactic chemical evolution and understanding the cool stars and exoplanets [1]. Besides, they are necessary to determine impurity concentrations from fusion plasma spectra [2], to develop new metal-halide high-intensity discharge lamps, to understand the crystal spectra of the divalent and trivalent salts [3].

Neutral gadolinium (Z=64), one of the rare earth elements, has an electronic structure of a half-full 4*f* electron shell and a 5*d* electron. It is complicated to calculate Gd I transition probabilities for the strong collapse of the 4*f* orbital and for the huge number of levels arising from the configurations involving an open 4f shell [3]. An experimental determination of these quantities, widely used in the present, is the combination of lifetime measurements using laser-induced fluorescence technique with branching fraction measurements using Fourier-transform or grating spectrometer. This method is more reliable than that by absorption or emission spectra since accurate atomic density is difficult to obtain. Therefore, lifetime measurements are of importance in obtaining transition probabilities.

Early lifetime measurements of Gd I were performed using the delayed coincidence method by Marek and Stahnke for levels between 17,380 and 27,337 cm^{-1} [4] and by Gorshkov et al. for levels between 22,334 and 29,452 cm^{-1} [5]. Later on, by a three-step delayed photoionization method, Miyabe et al. published Gd I lifetimes for levels in the 16,061– 36,361 cm^{-1} region [6]. More recently, using the time-resolved laser induced fluorescence technique, lifetime measurements of Gd I were reported by Xu et al. for levels ranging from 26,866 to 36,395 cm^{-1} [7], by Den Hartog et al. for levels from 17,750 to 36,654 cm^{-1} [8], by Feng et al. for levels from 29717.231 to 41692.155 cm^{-1} [9] and by Wang et al. for levels from 27014.751 to 38434.97 cm^{-1} [10].

Although a lot of efforts were devoted to the investigation of Gd I, their lifetime data are still fragmentary. In this paper, radiative lifetimes for 27 excited levels of Gd I were measured using TR-LIF technique in an atomic beam produced by laser-induced plasma.

2. Experimental setup

The experimental setup used in the present experiment is shown in Fig. 1. Free atoms were obtained by laser ablation on the gadolinium target foil which was placed in a vacuum chamber and rotated during the experiment. The ablation laser with 532 nm wavelength, 8 ns pulse duration and about 10 mJ energy, emitted by a Q-switched Nd:YAG laser working at 10 Hz repetition rate, was perpendicularly focused on the gadolinium target. In order to obtain the suitable excitation, 532 nm pulses with 8 ns duration, emitted by another Q-switched Nd:YAG laser working at 10 Hz repetition rate, were sent to pump a dye laser operating with the

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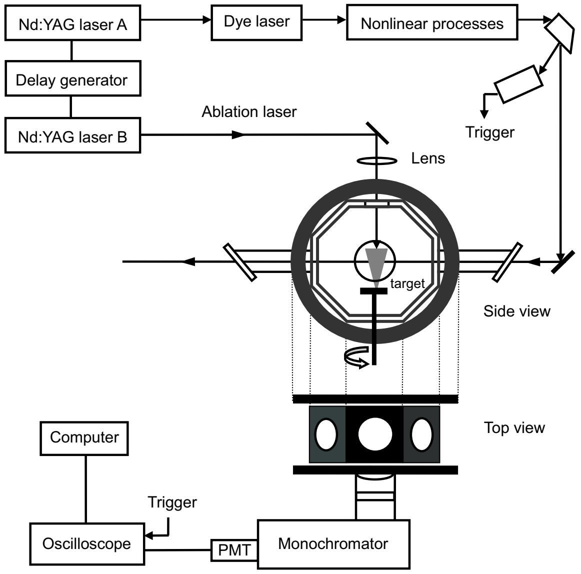

Fig. 1. Experimental setup used for lifetime measurements.

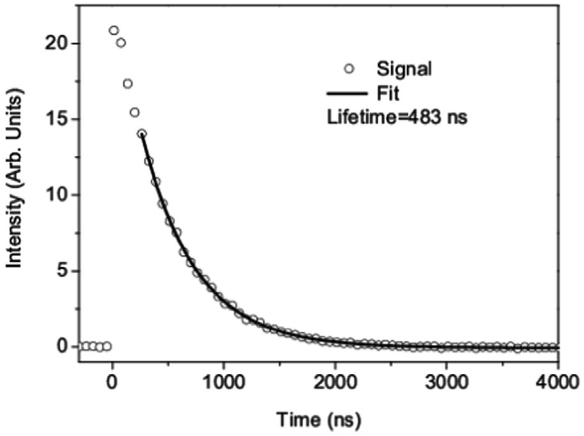

Fig. 2. Typical fluorescence decay curve of the 28486.854 cm^{-1} level of Gd I with an exponential fitting.

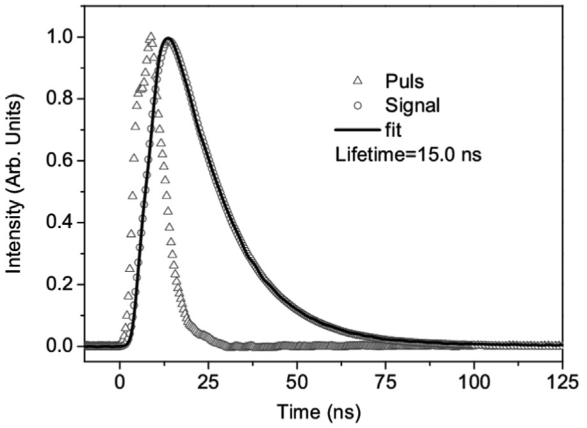

Fig. 3. Typical fluorescence decay curve of the 36482.515 cm^{-1} level of Gd I with a convolution fitting.

DCM dye. The frequency-doubling of the dye laser with an about 0.08 cm^{-1} line width was produced by a BBO type-I crystal, and then was focused into a hydrogen cell to obtain different orders of Stokes Raman-shifted components providing tunable radiation from 345 to 381 nm. Then the excitation laser was sent horizontally through the sputtered cloud of atoms at a distance of about 8 mm above the target surface.

The two Nd: YAG lasers were triggered by a digital delay generator which provided different time intervals between the ablation and excitation pulses. In this work, the delay was changed in a range from 10 to 102 μs for Gd I. The fluorescence decay signal emitted from the excited states under investigation was focused on a grating monochromator ($f = 10$ cm) by a fused silica lens in the direction perpendicular to the excitation laser and the atomic plume. The fluorescence signal selected by the monochromator was detected by a photomultiplier tube and recorded by a digital oscilloscope.

3. Lifetime measurements

In the measurement, one-photon excitation scheme was employed. To ensure that the upper level is correctly excited and free from blends, the excitation laser wavelength was carefully chosen from all available excitation pathways to avoid co-excitation and/or mis-excitation of other levels. Moreover, by verifying that all the fluorescence channels were related to the upper level and the lifetime value measured at different fluorescence channels were the same in error bars, the excitation of correct upper level can be confirmed. Also the decay channel with larger branch fraction was generally used for fluorescence detection.

In the experiment, several systematic effects may influence lifetime measurements. Radiation trapping effect will prolong the measured lifetime value when the density of excited atom is too large. This effect could be eliminated through depressing the pulse energies of the excitation and ablation lasers and increasing the delay time between the ablation and excitation pulses. Flight-out-of-view effect is important for long lifetime measurements (>1 μs) when excited atoms have larger speeds. This effect could be effectively eliminated by increasing the delay time to obtain lower-speed atoms and widening the slit width of the monochromator to expand the fluorescence collection area. The effect of collision-induced quenching caused by collisions with remnant gas can be removed by reducing the pressure in the vacuum chamber to below 0.3×10^{-3} Pa. The excitation laser intensity may be properly weakened to avoid saturation effect. If fluorescence signal was strong enough, the PMT voltage may be turned down to avoid nonlinear response of photoelectric signal. A magnetic field of about 100G produced by a pair of Helmholtz coils was applied in the direction of the horizontal component of the earth's magnetic field. It may not only eliminate possible quantum beats due to the Earth's field but also weaken the recombination background in plasma [11]. The background intensity varies slowly compared with the fluorescence decay time. So, for short-lived levels shorter than 100 ns this background can be neglected. For long-lived levels, the background was recorded separately without the excitation pulse and subtracted from the fluorescence signal.

To improve the signal-to-noise ratio, more than 1000 shots were averaged for each fluorescence decay curve. The lifetime values longer than 50 ns were obtained by fitting the recorded fluorescence curve to an exponential function with adjustable parameters. A typical decay curve of the 28486.854 cm^{-1} level in Gd I with an exponential fitting is shown in Fig. 2. For the lifetime shorter than 50 ns, the excitation laser pulse was recorded by the same detection system. By fitting the fluorescence signal to the convolution of the detected laser pulse and a pure exponential function, the lifetime values were obtained. In this process the effects of the finite duration of the excitation laser pulse and the limited response time of detection system can be well eliminated. As an example, the fluorescence decay curve of the level 36482.515 cm^{-1} of Gd I, the recorded excitation laser pulse shape