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Measurement of total angular momentum values of high-lying even-parity atomic states of samarium by spectrally resolved laser-induced fluorescence technique

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Abstract. Spectrally resolved laser-induced fluorescence technique was used to uniquely assign total angular momentum (J) values to high-lying even-parity energy levels of atomic samarium. Unique J value assignment was done for seven energy levels in the energy region 34,800–36,200 cm^{-1}, recently observed and reported in the literature.

Keywords. Laser spectroscopy; laser-induced fluorescence; samarium.

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1. Introduction

Knowledge and information of high-lying energy levels are important for atomic structure calculations and to choose efficient photoionization schemes for trace and ultra-trace analyses. The study of high-lying energy levels in the intermediate energy region could be possible only with the advent of tunable lasers, which were then selectively, and appreciably populated using different excitation sources and pathways. Total angular momentum value (J) is fundamentally important in choosing possible excitation schemes and, hence, assignment of its value to each energy level of the excitation pathways is of great significance. There are several experimental methods to determine J of a given atomic energy level, e.g., Zeeman effect, hyperfine splitting in absorption lines, spectrally resolved fluorescence from the level, excitation to same level via multiple excitation pathways and polarization combination methods. Zeeman effect and hyperfine splitting in absorption lines [1] can be applied to ground or low-lying atomic energy levels, which can be populated by single excitation source. Multiple excitation pathways [2,3] or spectrally resolved fluorescence technique [4,5] can be successfully employed for assigning a unique J value to high-lying energy levels. Identification of J for autoionizing energy levels uses polarization combination method [6] as all the other above-mentioned methods become very cumbersome and difficult to apply.

Samarium is a rare-earth element and the study of its energy levels is significant due to effects of parity non-conservation, which are expected to be of higher order for samarium. The limited knowledge of even-parity spectrum in the higher energy region in the literature has motivated in the recent past for new energy level search and for determination of J values of the levels in this energy region. Jayasekharan *et al* [3] used the method of multiple excitation pathways to identify eighty-six even-parity levels of Sm I in the energy region 32,950–36,000 cm^{-1} and uniquely assigned J values to most of them. Jia *et al* [5] resolved laser-induced fluorescence spectrally with a monochromator and could partially ascertain J values to high-lying even-parity energy levels of Sm I. In this paper, we have spectrally resolved two-colour laser-induced fluorescence from the excited high-lying energy level of atomic samarium and assigned unique J values to seven such energy levels in the energy range 34,800–36,200 cm^{-1} from the observed fluorescence decay routes to lower energy levels.

2. Experimental method

Tunable narrow band lasers to selectively excite high-lying energy levels and sensitive detection systems to detect weak atomic fluorescence spread over wide spectral regions have enabled us to study energy regions which were inaccessible with conventional techniques. We have employed tunable dye lasers (Quantel TDL90) pumped by a Q-switched Nd:YAG laser (Quantel YG980) for resonant photoexcitation to chosen high-lying energy levels in atomic samarium. The experimental set-up used for this study is shown in figure 1. The details of our LIF experimental chamber along with samarium vapour generation and fluorescence collection assembly are given in our earlier paper [2]. The samarium atomic beam was generated effusively in a vacuum chamber by resistive heating of samarium metal. The fluorescence light emitted from the excited upper levels was collected perpendicularly to both laser and atomic beams and focussed onto the entrance slit of the 0.5 m monochromator (Acton SpectraPro, 2500i) by a suitable lens assembly. The monochromator consists of 500-nm blazed grating having 300 and 1200 lines/mm. Wavelength/pixel calibration of the spectrograph was performed using Hg/Ar lamp. For the grating having 300 lines/mm, the covered wavelength range was 130 nm for single coverage. So, multiple acquisitions have been carried out to cover the LIF spectrum in the wavelength range 300–750 nm. The wavelengths for the first- and second-step excitations were the same as used in our earlier work [2], where wavelength calibration was done using a U–Ne hollow cathode lamp and Fabry–Perot etalon fringes.

For observation and optimization of two-colour LIF, single-colour LIF signal was first optimized and the second laser was tuned for maximum two-colour LIF signal. After optimization, LIF signal was directed towards the ICCD camera. It displayed all resonant and non-resonant decay channels within the spectral range of the monochromator. The decay channels due to the excitation by two-photons of the same second excitation laser were identified by blocking the first excitation laser. These decay channels were ignored from the list of decay channels from the

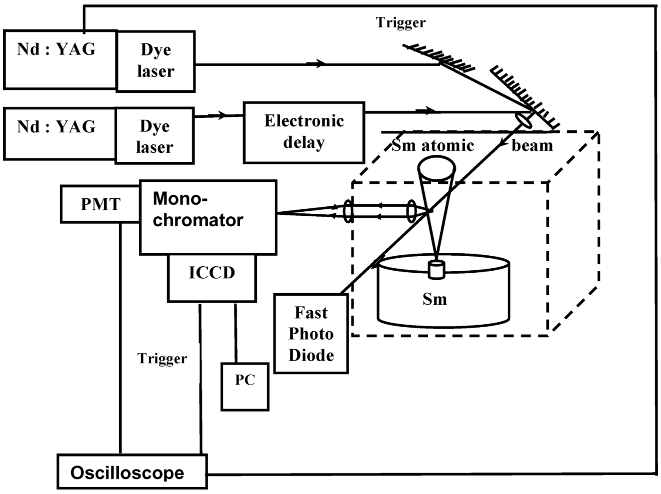

Figure 1. Experimental set-up for spectrally resolved two-colour laser-induced fluorescence.

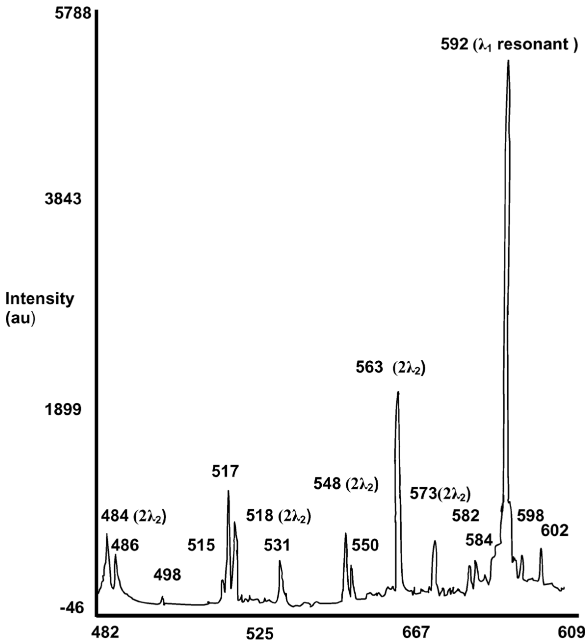

Figure 2. Spectrally resolved laser-induced fluorescence decay channels from step-wise excited even-parity energy level at 34935.5 cm^{-1} using two-step excitation ($\lambda_1 = 591.64$ nm, $\lambda_2 = 563.36$ nm).

two-colour step-wise excited levels. All the decay channels from a step-wise excited energy level were tabulated and their respective low-lying energy level was assigned with the help of energy level information available in [7]. Unique identification of

Table 1. Observed fluorescence decay channels from the step-wise excited even-parity energy level at 34935.5 cm^{-1} to various lower odd-parity levels.

Fluorescence wavelength (nm)	Energy of lower level (cm^{-1})	J value of lower level
486.3	14380.50	2
498.1	14863.85	1
514.6	14915.83	3
516.1	15567.32	2
531.2	16116.42	2
549.7	16748.30	3
582.4	17769.71	1
584.4	17830.80	3
597.7	18209.04	3
602.0	18328.64	3

Table 2. Assigned J values to seven step-wise excited high-lying even-parity energy levels of atomic samarium along with the used excitation wavelengths.

Energy level (cm^{-1})	First-step excitation wavelength (λ_1) (nm)	Second-step excitation wavelength (λ_2) (nm)	Total angular momentum value (J)
34814.4	591.64	567.22	2
34924.0	591.64	563.73	2
34935.5	591.64	563.36	2
34972.1	591.64	562.20	3
35072.6	591.64	559.04	1
35092.1	591.64	558.44	3
36188.0	570.62	581.95	2

the total angular momentum value was done using electric dipole selection rules $\Delta J = 0, \pm 1$.

3. Results and discussion

High-lying even-parity energy levels of samarium atoms were selectively populated by two-step laser excitation. Fluorescence from these levels was spectrally resolved. Lower odd-parity energy levels to which two-step excited atoms decay were identified for the observed fluorescence decay channels in each case using the energy level information available in the literature [7]. Figure 2 shows typically a portion of the spectrally resolved scan of radiative decay from 34935.5 cm^{-1} even-parity energy level. Similar scans of LIF decay from other high-lying even-parity energy levels were recorded. Table 1 lists the observed fluorescence decay channels from the step-wise excited even-parity energy level at 34935.5 cm^{-1} to various lower odd-parity levels. The fluorescence wavelengths, energies and J values of the lower levels to which the excited level decays are tabulated. Fluorescence decay channels, which are only due to two-step resonant excitation, are used for ascertaining J value of the upper level. From table 1 and similar tables (not given) for other high-lying even-parity energy levels, we have inferred the J values of the seven even-parity energy levels based on examining J values of the lower odd-parity levels to which a stepwise-excited even-parity level decays. Table 2 lists the high-lying even-parity energy levels along with the excitation wavelengths and the assigned J values.

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