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Measurement of radiative lifetime in atomic samarium using simultaneous detection of laser-induced fluorescence and photoionization signals

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Abstract. In this paper, we report the investigations of lifetime measurement of odd-parity energy level 19009.52 cm^{-1} of Sm I using simultaneous detection of laser-induced fluorescence and laser-induced photoionization signals employing pump–probe technique. To the best of our knowledge, this is for the first time that the results obtained using laser-induced fluorescence and photoionization techniques have been compared with each other. The obtained results match well with those reported in the literature.

Keywords. Laser-induced fluorescence; laser-induced photoionization; radiative lifetime; samarium.

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1. Introduction

Knowledge of accurate atomic data of lanthanide elements is useful for the development of laser technology, investigations on atomic, molecular and plasma physics, laser chemistry and to check accuracy of theoretical atomic models. Moreover, the data are of importance in astrophysics as these elements have cosmic abundance and thus rich in stellar spectral lines. Advent of pulsed tunable dye laser has proved to be an excellent tool for determining atomic radiative lifetimes [1–4]. In this paper, we report the investigations of radiative lifetime measurement of odd-parity energy level at 19009.52 cm^{-1} of Sm I using simultaneous detection of laser-induced fluorescence (LIF) and laser-induced photoionization (LIP) signals employing the pump–probe technique. To the best of our knowledge, this is for the first time that the results obtained using LIF and LIP techniques have been compared with each other as well as with those reported in the literature.

2. Experimental

An atomic beam of samarium was produced in a vacuum chamber maintained at a background pressure of about 10^{-5} torr. The oven consists of a hollow tantalum cylinder with a lid having a 0.5 mm hole. The atomic beam of samarium was produced by resistively heating a tantalum-heating element around a tantalum crucible containing samarium metal to ∼1000°C. Samarium atoms effuse out from the orifice in the form of an atomic beam. The atomic beam was irradiated with Nd-YAG pumped dye laser beams at the centre of atomic beam chamber. The pulsed laser was operated with a repetition rate of 20 Hz and a pulse width of ∼7 ns. The first laser used as pump laser was tuned to a resonant wavelength at 570.62 nm to excite the atoms in the ground-state septet to the intermediate odd-parity energy level at 19009.52 cm^{-1}, whose radiative lifetime measurement is to be taken. Non-resonant fluorescence signal from the first-step excited level was observed to attain the first-step resonance and the laser was kept at the maximum fluorescence signal. Fluorescence light was collected with two-lens assembly each of focal length of 20 cm (distance from interaction zone to window port) and diameter of 10 cm. The second lens focusses the collected light on the entrance slit of 0.5 m monochromator (Acton SpectraPro, 2500i), which consists of three interchangeable 500 nm blazed gratings having 300, 600 and 1200 lines/mm. The resolution of the spectrograph with 300 lines/mm grating is roughly 0.1 nm, which has been used in this study. At the exit slit of the monochromator, the photomultiplier tube (PMT) with a gain of $\sim 10^7$ is coupled whose output is fed to an oscilloscope. The PMT detects the sum of fluorescence light emitted within the band pass of the monochromator. The second-step laser was tuned across the tuning range of the dye to find the second-step resonant wavelengths. The probe laser at one of these resonant second-step wavelengths further excites the atoms in the intermediate step to a high-lying, even-parity level. To improve the excitation efficiency, both the dye lasers were ensured to be spatially overlapped and temporally synchronized. The monochromator position was tuned to achieve maximum second-step non-resonant fluorescence signal and minimum scattered background. The detection of non-resonant signal cuts off the contributions from the resonance fluorescence at excitation wavelength and laser scattering from the windows. The monochromator was set at a position such that it can cut down the laser scattering from both the lasers and pass the non-resonant fluorescence within its band pass.

The photoion signal was detected simultaneously using a parallel-plate configuration and applying −1 kV to the plates across a load of 5 kΩ. The photoion detection system was in cross configuration with laser and atomic beams, similar to the fluorescence collection assembly. As the fluorescence assembly was in a straight line with the photoion system, one of the plates was made of fine wire mesh with light transmission efficiency of about 80% so that maximum fluorescence light passed through it and reached the fluorescence collection assembly. The typical pulse energy of both dye lasers was about a few hundreds of microjoule and the laser linewidth was ~ 0.05 cm^{-1}.

The non-resonant two-colour fluorescence and two-colour three-photon photoionization signals were simultaneously monitored as a function of delay between the pump and the probe lasers. Temporal delay between the pump and the probe lasers was

introduced by an electronic delay generator between the two Q-switch pulses of Nd-YAG laser. The slope of the fluorescence and photoionization decay curves plotted against the delay between the pump and probe lasers provided radiative lifetime of the intermediate level.

3. Results and discussion

To establish the versatility of the technique and reliability of our experimental set-up, an attempt was made to measure the radiative lifetime of odd-parity energy level at 19009.52 cm^{-1}. We used the first-step excitation wavelength $\lambda_1 = 570.62$ nm corresponding to atomic transition (1489.55 $cm^{-1} \rightarrow$ 19009.52 cm^{-1}) to populate the 19009.52 cm^{-1} energy level. The second-step laser (probe laser) tuned to the wavelength $\lambda_2 = 566.89$ nm was used to further excite the atoms to high-lying, even-parity level. To improve the signal-to-noise ratio and to reduce the effect of pulse-to-pulse laser fluctuation, the average of 128 laser pulses was taken. Laser-induced fluorescence and laser-induced photoionization decay plots for this level as the probe laser was temporally delayed are shown in figures 1 and 2, respectively.

The measurements were repeated with 30% of the laser power used in the previous experimental observation. We have not observed any significant change in the lifetime values. The measurements were again repeated with higher oven temperature to check the effect of self-absorption but the lifetime values obtained were within the experimental

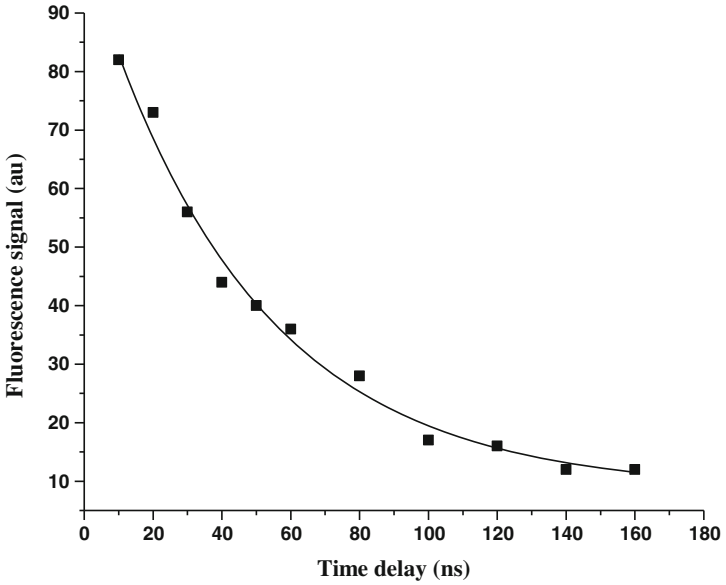

Figure 1. Laser-induced fluorescence decay curve for the energy level 19009.52 cm^{-1}.

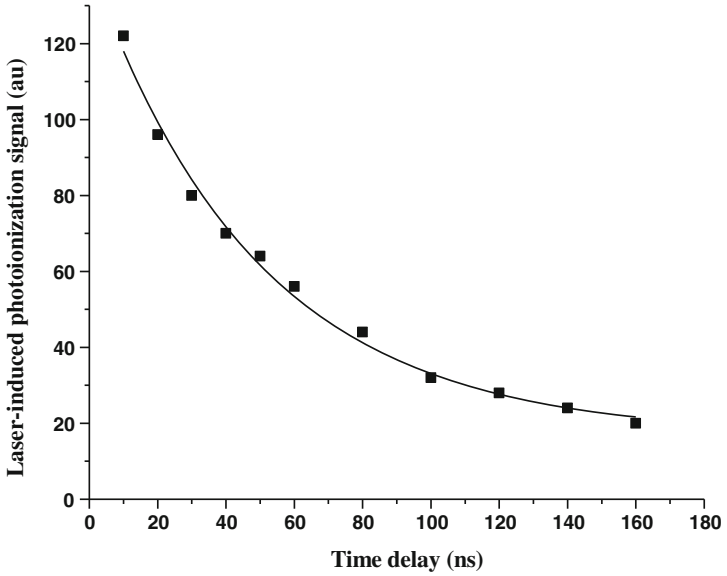

Figure 2. Laser-induced photoionization decay curve for the energy level 19009.52 cm^{-1}.

errors. These experiments were repeated with a different probe laser at 568.57 nm. The results are shown in table 1 along with the values reported in [4,5].

The results obtained using laser-induced fluorescence and photoionization match well with each other and with those reported in the literature. The lifetime values given in table 1 are average of several measurements, but we obtain the final lifetime value of the odd-parity energy level 19009.52 cm^{-1} of Sm I as 47 $\pm$ 4 ns based on the average of the lifetime values mentioned in table 1.

Table 1. Lifetime values for the odd-parity energy level 19009.52 cm^{-1} of Sm I using simultaneous laser-induced fluorescence and photoionization.

Methodology	Lifetime value with laser-induced fluorescence (ns)	Lifetime value with laser-induced photoionization (ns)	Lifetime value reported in the literature [4,5] (ns)
Using probe laser at 566.89 nm	47	49	46 and 51
When laser power was attenuated to 30%	48	47	
Oven temperature increased by 100°C	47	48	
Using probe laser at 568.57 nm	46	47	

4. Uncertainties in measurements

The statistical uncertainty in our measurements of the lifetime is $\sim\pm10\%$ based on the spread in the lifetime values measured several times. Apart from this statistical uncertainty, there could be various effects causing systematic uncertainties in the lifetime measurements, such as radiation trapping, collisional depopulation, atomic motion, cascade repopulation and depopulation, Zeeman and hyperfine quantum beats [6]. The radiation trapping is insignificant in our experiments carried out in a Sm atomic beam with the number density in the laser-interaction zone estimated to be 10^{11} atoms/cm^3. We have not observed any change in the measured values of lifetime even at higher permissible oven temperatures or number density as also evident from table 1. The collisional depopulation of excited levels is also insignificant in the present work as experimental measurements have been carried out in a vacuum chamber evacuated to a pressure of 10^{-5} torr. We have not found the evidence of collisional quenching even at a pressure of 10^{-4} torr. The atomic motion effect can shorten the lifetime because the radiative atoms can move out into a region of lower collection efficiency. The effect is negligible in the case of short-lived levels with lifetime less than 200 ns as the transit time of atoms is a few microseconds. As our measured lifetime value is less than 200 ns, the systematic error due to this effect is negligible. The selective excitation of atomic energy levels using lasers eliminates the possibility of cascade repopulation from higher-lying levels. There could be some modulations in the observed fluorescence intensity due to Zeeman and hyperfine quantum beats. The residual magnetic field in the interaction zone due to current flowing in the heater for the generation of Sm atomic beam was very small ($\sim$ 60 mG). The natural abundance of odd Sm isotopes is about $\sim$28%, so only 28% of the total fluorescent and photoionization signal can be modulated due to hyperfine quantum beats. We have not observed any modulation in the fluorescence and photoionization decay curves due to these effects. Our values of the lifetimes are free from the common systematic errors. Thus, the systematic uncertainties have been neglected because any small contribution possible due to systematic errors would not change the uncertainty beyond a statistical value of 10%.

5. Conclusion

We have measured the radiative lifetime of the odd-parity energy level 19009.52 cm^{-1} of Sm I using simultaneous laser-induced fluorescence and photoionization signals by employing the pump–probe technique. Our emphasis was not to generate volume of new data but to compare laser-induced fluorescence and photoionization techniques for such measurements. The lifetime values obtained using laser-induced fluorescence and photoionization techniques match well with each other and with those reported in the literature. To the best of our knowledge, this is the first that laser-induced fluorescence and photoionization techniques are used simultaneously to measure lifetime and the results are compared with each other as well as with other reported values. The technique used here can be universally applicable for other energy levels of interest as well as for other materials.

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