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Measurement of photoexcitation cross-sections of uranium by saturation method

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Abstract. We report the measurement of photoexcitation cross-sections of three first-step uranium transitions ($0 \rightarrow 16900.38$ cm^{-1}, $0 \rightarrow 17361.89$ cm^{-1} and $620 \rightarrow 17361.89$ cm^{-1}) using saturation method. These measurements were performed on a resonance ionization mass spectrometry (RIMS) set-up consisting of Nd:YAG-pumped dye lasers, a reflectron time-of-flight mass spectrometer and high-temperature atomic vapour source. The uranium vapours were excited and photoionized by two-colour, three-photon photoionization scheme using Nd:YAG-pumped dye laser system. The resultant photoion signal was monitored as a function of dye laser fluence used for first step excitation to measure the excitation cross-section values. A new approach was adopted to overcome the large uncertainties associated with such measurements. With this approach the cross-section of transitions whose value is already reported in the literature was measured as a bench mark. By normalizing the measured value to the reported value, a scaling factor was derived. This scaling factor was used to scale up the cross-section values of other transitions measured by this method.

Keywords. Photoexcitation cross-section; resonance ionization mass spectrometry; time-of-flight mass spectrometer; uranium.

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1. Introduction

The knowledge of photoexcitation/photoionization cross-section is crucial in many areas of basic science, such as atomic physics, plasma physics, atmospheric sciences, for determination of solar and stellar abundances of elements, for searching the new laser materials, etc. The accurate value of this parameter is also very important for choosing efficient photoionization schemes for atomic vapour laser isotope separation process, trace analysis etc. as it determines the laser fluences needed for each step involved in the phoionization process.

Commonly employed techniques for measuring this parameter rely on measurements of line strengths from absorption and emission spectra, measurement of saturation energy, Rabi frequency and the lifetime–branching-ratio [1,2]. We report the measurement of photoexcitation cross-sections of a few first-step

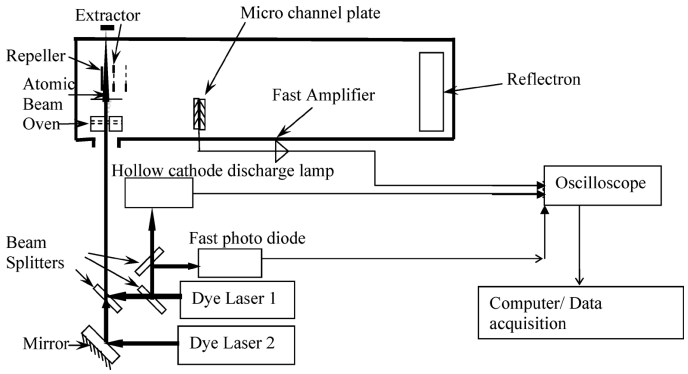

Figure 1. Experimental set-up.

uranium transitions starting from ground/metastable states ($0 \rightarrow 16900.38$ cm^{-1}), ($0 \rightarrow 17361.89$ cm^{-1}) and ($620 \rightarrow 17361.89$ cm^{-1}) using saturation method.

2. Experimental details

The experimental set-up is shown in figure 1, which consists of Nd:YAG laser (Model YG 980) pumped dye lasers (Model TDL 90), indigenously developed reflectron-type time-of-flight mass spectrometer, digital oscilloscope, home-made hollow cathode discharge lamp and a fast photodiode. Uranium atomic beam was generated by resistive heating of uranium metal in tantalum crucible in a vacuum chamber having vacuum better than 10^{-7}mbar. Uranium vapours, thus produced, were excited and photoionized by two-colour, three-photon photoionization scheme (one photon of the first laser and two photons of the second laser) using Nd:YAG-pumped dye laser system. The resulting photoion signal was detected using microchannel plate (MCP) in a reflectron-type time-of-flight mass spectrometer. This signal was monitored on a digital oscilloscope as a function of dye laser fluence used for the first step excitation. Spatial overlap of both the dye laser beams with each other and with atomic beam in the interaction zone was ensured. A delay of $\sim$10 ns was kept between the first and second step lasers to ensure sequential excitation and to avoid the effect of second laser on the saturation due to the first laser. First-step laser wavelength acquisition was done by observing optogalvanic signal in home-made U–Ne hollow cathode discharge tube. Second laser was tuned for further excitation and ionization.

The precise value of absolute cross-section depends on the accurate measurement of laser and atomic parameters such as laser spot size in the interaction zone, laser power, laser linewidth, atomic absorption linewidth, spatial overlap of both the laser beams with themselves and with atomic beam etc. To take care of the inaccuracies arising in these measurements, a new approach was adopted. In this approach, the cross-section of transition whose value is already reported in the literature was measured. By normalizing the measured value to the reported value, a scaling factor was derived. This scaling factor was used to scale up the cross-section value of other transitions measured by this method. To implement this approach, we have

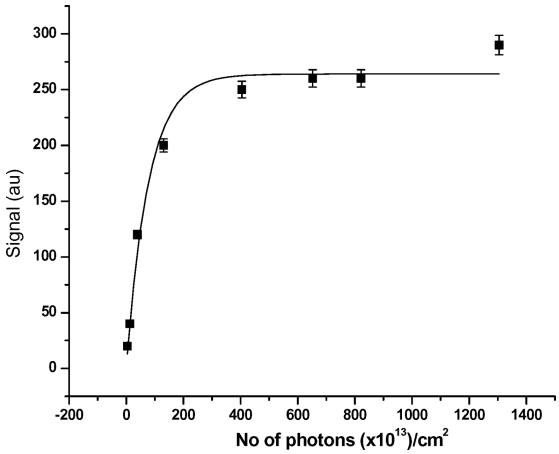

Figure 2. Saturation curve for $0 \rightarrow 16900.38$ cm^{-1} transition.

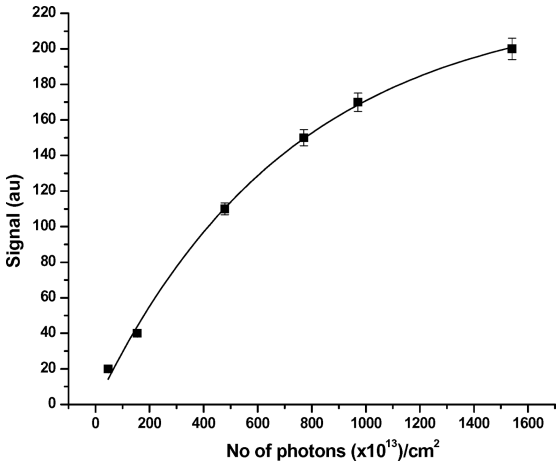

Figure 3. Saturation curve for $0 \rightarrow 17361.89$ cm^{-1} transition.

chosen first-step transition of uranium ($0 \rightarrow 16900.38$ cm^{-1}), at a laser wavelength of 591.5 nm whose cross-section value reported in the literature is 1.1×10^{-13} cm^2 [3]. We have measured the cross-section value for this transition by monitoring the photoionization signal as a function of dye laser fluence used for the first-step excitation.

Figure 2 shows the saturation curve for this ($0 \rightarrow 16900.38$ cm^{-1}) uranium transition. The cross-section value was measured from this curve and a scaling factor was obtained from these two values. This scaling factor was cross-checked by measuring the cross-section value of the other uranium transition ($0 \rightarrow 17361.89$ cm^{-1}), whose cross-section is known. Figure 3 shows the saturation curves for this uranium transition. The cross-section value was obtained by multiplying with the scaling factor to give the actual cross-section value of 1.4×10^{-14} cm^2, which was

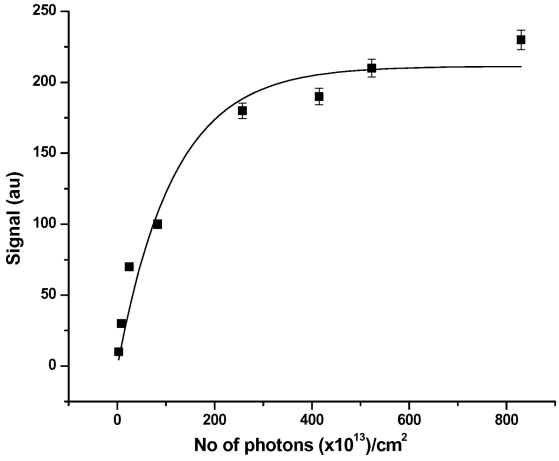

Figure 4. Saturation curve for 620 → 17361.89 cm^{-1} transition.

found to be in good agreement with that reported in the literature (1.6×10^{-14} cm^2) [3], thus validating our scaling factor. Finally, we measured the cross-section value of a transition starting from 620 cm^{-1} metastable state (620 → 17361.89 cm^{-1}), whose cross-section value is not known. Figure 4 shows the saturation curves for this transition and the measured value after scaling it up is 6.8×10^{-14} cm^2.

3. Conclusion

To compensate for all the errors involved in the conventional saturation method for the measurement of excitation cross-section, we have obtained a scaling factor from the cross-section measurement of a known transition and used it to measure the cross-sections for other transitions. The measured values after using the scaling factor were found to be in good agreement with those reported in the literature. This approach can thus be applied to other transitions where no data are reported. We have planned to extend this approach to measure the second and third step transition cross-section values where either data are not available or not published.

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