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# Photoionization cross-section measurements from the 2p, 3d and 3s excited states of lithium

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**Abstract.** The photoionization cross-sections from the $2p\ ^2P_{1/2,3/2}$, $3d\ ^2D_{3/2,5/2}$ and $3s\ ^2S_{1/2}$ excited states of lithium have been measured at different ionizing laser wavelengths, above the first ionization threshold. The experiments are performed by using a thermionic diode working in the space charge limited mode and the cross-sections are measured by employing the saturation technique. By changing the ionization photon energy, a smooth frequency dependence of the cross-sections has been observed for the $2p$ and $3d$ states. The cross-section from the $3s$ excited state has been measured at a single photon energy. The measured values of the photoionization cross are compared with the available data.

**PACS.** 32.30.Jc Visible and ultraviolet spectra – 32.80.Rm Multiphoton ionization and excitation to highly excited states

## 1 Introduction

Lithium possessing a single valence electron is the simplest element after hydrogen and helium. The Rydberg series of lithium to higher principal quantum numbers and an accurate value of the ionization potential have been studied systematically by many groups [1–3] but comparatively little information is available about the cross-sections of its excited states. Rothe [4] pioneered studies of the photoionization cross-section for the excited states of Li based on studies of the radiative electron-ion recombination and shock-heated plasma. The photoionization cross-section at threshold was measured by using absolute intensity of the signal. The atomic beam and laser saturation technique enabled measurement of the photoionization cross-sections of the excited alkali atoms [5–10]. Aymar et al. [7] computed the photoionization cross-section for the $s$, $p$ and $d$ Rydberg states of Li in the framework of a single-electron model by using a parametric central potential and compared the results with those calculated using quantum defect theory [11]. Kramer et al. [12] reported the resonant three photon ionization process of lithium and measured the photoionization cross-section at 610.4 nm using the saturation technique. Barrientos and Martin [13] applied the quantum defect orbital method to compute the oscillator strengths and the photoionization cross-sections of lithium; continuity across the series limit revealed the cross-section at the ionization-threshold. Lahiri and Manson [14] calculated the radiative-recombination rate coefficients for electrons impinging on $Li^+$, along with the associated excited-state photoionization cross-sections in the low-energy region. A brief review of theory and the relationship between the photoionization and radiative-recombination was also presented. Chung [15] calculated the photoionization cross-section for Li $^2S$ and $^2D$ states from the ionization threshold to the $Li^+$ and predicted the even-parity Li resonances below the $Li^+$ threshold.

Magneto optical traps (MOT) have also been successfully used for measuring the absolute photoionization cross-sections of the excited alkali atoms. This technique was first introduced by Dinneen et al. [16], who pioneered the MOT loaded by an atomic beam to measure the photoionization cross-section of the $5p\ ^2P_{3/2}$ state of rubidium. Wippel et al. [17] reported the photoionization cross-sections of the first excited states of the two isotopes of lithium ($^7$Li and $^6$Li) with different ionizing lasers using a magneto-optical trap. A fraction of the trapped atoms in the first excited state was ionized using the UV-laser radiation. With the help of the loading curve, they determined the photoionization cross-sections of the $2p$ excited state of lithium at 334.47 nm and 335.85 nm above the first ionization threshold. Recently, we reported the photoionization cross-sections of the $3p\ ^2P_{1/2}$ and $^2P_{3/2}$ excited states of sodium at 355 nm above first the ionization threshold using the saturation technique [18].

The present paper describes the measurements of the photoionization cross-sections from the $2p$, $3d$ and $3s$ states of lithium. The $3d$ and $3s$ states are populated by the two-photon excitation from the ground state.

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The experimentally measured cross-sections from the aforementioned states as a function of laser frequency are reported for the first time.

## 2 Experimental set-up

The basic experimental arrangement to measure the photoionization cross-section from the excited states of lithium is similar to that described in our earlier work [3,18,19]. The laser system comprised a Q-switched pulsed Nd:YAG laser (Quantel, Brilliant), 10 Hz repetition rate, pulse duration ≈4 ns, energy 180 mJ per pulse at 532 nm. The line width of the laser is 1.4 $cm^{-1}$ at 532 nm. The output of the SHG of the Nd:YAG laser was used to pump two dye lasers, a commercial (Quanta-Ray PDL-2) and a home made Hanna type Dye laser cavity [20]. The cavity was formed between a flat mirror and a 2400 lines/mm holographic grating and wavelength tuning was achieved by rotating the grating by a computer controlled stepper motor. The lithium vapors were produced in a thermionic diode ion detector composed of a stainless steel tube 48 cm long, 3 cm in diameter and 1 mm wall thickness. About 20 cm of the central part of the tube was heated by a clamp-shell oven operating at a temperature 825 K that corresponds to about 0.02 Torr lithium vapor pressure. Argon gas at a pressure of ≈0.5 Torr was used as a buffer gas, which provides a uniform column of the lithium vapors. The temperature was monitored by a Ni–Cr–Ni thermocouple and it was maintained within ±1% by a temperature controller. A molybdenum wire 0.25 mm in diameter stretched axially was heated by a separate regulated power supply that served as cathode for the ion detection.

The photoionization cross-sections from the $2p$, $3d$ and $3s$ excited states of lithium were measured as a function of the ionizing laser frequency. In order to measure the photoionization cross-section from the $2p$ excited state, the atoms in the first step were resonantly excited from the $2s\ ^2S_{1/2}$ ground state to the $2p\ ^2P_{1/2,3/2}$ state (670.9 nm) using a dye laser, charged with LDS 698 dye, dissolved in methanol and pumped by the second harmonic of a Nd:YAG laser. In the second step, these excited atoms were ionized using a laser at 349.8 nm, 335 nm and 266 nm. A BBO crystal was used for the frequency doubling to reach the aforementioned wavelengths. In each experiment the diameter of the exciting laser was ≈1 mm and the cross-sectional areas of the ionizing lasers were calculated for each wavelength. In these experiments the dye laser pulse energy was ≈1 mJ and bandwidth ≤0.3 $cm^{-1}$. The ionization signal was optimized by monitoring the cathode current and the buffer gas pressure. The intensity of the ionizing laser was varied by inserting the neutral density filters (Edmund Optics) and on each insertion its energy was measured by an energy meter (R-752, Universal Radiometer). The ion signal was taken across a 10 kΩ resistor through a 0.01 μF blocking capacitor and was registered on a 100 MHz storage oscilloscope.

In the second set of the experiments we measured the photoionization cross-section from the $3d$ excited state. The first dye laser (Quanta-Ray, PDL-2) charged with the Rhodamine 640, dissolved in methanol and pumped by the SHG (532 nm) of the Nd:YAG laser was used to excite the atoms from the $2s\ ^2S_{1/2}$ ground state to the $3d\ ^2D_{3/2,5/2}$ states lying at 639.3 nm. This state is subsequently ionized by a second laser of variable frequencies and pulse energies. In each experiment the cross-sectional area was calculated, the intensity of the ionizing laser was varied by inserting the neutral density filters, and its energy was measured and the ion signal was registered on a storage oscilloscope. Similarly, we measured the photoionization cross-sections from the $3s\ ^2S_{1/2}$ excited state of lithium by using the two-photon excitation from the ground state. The $3s\ ^2S_{1/2}$ state was populated via two-photon excitation by tuning the dye laser to 735.1 nm. The second dye laser was adjusted to 613 nm to achieve ionization from this state.

## 3 Results and discussion

Alkali metals in their ground state configuration have a single electron in the $ns$ orbital and a core of inner electrons. The inner core has an overall spherical charge distribution and for the most part, the outer electron is likely to be found outside this core. This outermost electron is responsible for the transitions between different atomic states. Lithium is the first element of the alkali metals group possessing a single $s$-electron outside the filled $1s$ sub-shell. The excited states $ns\ ^2S_{1/2}$ , $np\ ^2P_{1/2,3/2}$, $nd\ ^2D_{3/2,5/2}$ and $nf\ ^2F_{5/2,7/2}$ can be approached via single photon, two-photon or three-photon excitation from the ground state. Measurements of the absolute photoionization cross-section of the excited states of alkali atoms have been extensively studied both theoretically and experimentally ([4–18] and references therein). The probability that a photon will produce ionization is a measure of the photoionization cross-section, which is obtained from the laser energy at which the photoionization process saturates. In the present work the absolute photoionization cross-sections above the first ionization threshold are determined using the saturation technique [21–24]. Burkhardt et al. [21] applied the two-step ionization technique to measure the absolute cross-section from the excited states of sodium, potassium and barium. These experiments were performed under the assumption that the intensity of the ionizing laser is higher i.e. in excess of that required for saturating the resonance transitions, and that the spontaneous emission is ignored during the 10 ns laser pulse. In addition it was assumed that the transitions remain saturated during the laser pulse and the laser beam is uniform and linearly polarized. Under these assumptions the total charge per pulse is given by

$$Q = eN_0V_{vol}\left[1 - \exp\left(-\frac{\sigma U}{2\hbar\omega A}\right)\right] \quad (1)$$

where $e$ (Coulomb) is the electronic charge, $N_0$ ($cm^{-3}$) is the density of excited atoms, $A$ ($cm^2$) is the cross-sectional area of the ionizing laser beam, $U$ (Joule) is the total

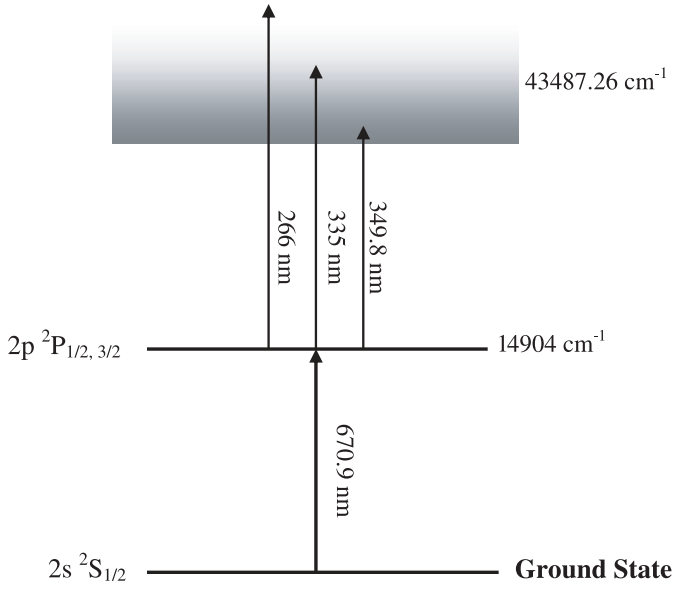


**Fig. 1.** Energy level diagram for the measurement of photoionization cross-sections from $2p$ excited states. The level energies are taken from NIST data.

energy per ionizing laser pulse, $\hbar\omega$ (Joule) is the energy per photon of the ionizing laser beam, $V_{vol}$ ($\mathrm{cm}^3$) is the laser interaction volume and $\sigma$ ($\mathrm{cm}^2$) is the absolute cross-section for photoionization. Subsequently, He et al. [22] reported the absolute photoionization cross-section of the $6s6p\ ^1\mathrm{P}_1$ excited state of barium. Their method for determining the photoionization cross-section is an extension of the saturation technique described by Burkhardt et al. [21] and accounting for the effects of the Gaussian laser intensity distribution. Mende et al. [23] modified equation (1) and acquired the photoionization cross-section by using the equation:

$$n_{ion} = N_{(level)} \int_V \left[1 - \exp\left(-\frac{1}{2}\sigma_{1\varepsilon}\phi_{ph}\right)\right] dV. \quad (2)$$

Here $n_{ion}$ is the number of photo ions, $\sigma_{1\varepsilon}$ is the cross-section for the photoionization and $\phi_{ph}$ is the time-integrated number of photons per unit area.

In the first experiment we have measured the photoionization cross-section from the $2p\ ^2\mathrm{P}_{1/2,3/2}$ state using a thermionic diode ion detector system. A schematic diagram of the pertinent transitions is shown in Figure 1. The dye laser at 670.9 nm is used to promote the atoms from the ground state to the $2p\ ^2\mathrm{P}_{1/2,3/2}$ states. The laser line width ($\sim$0.3 $\mathrm{cm}^{-1}$) of our system is nearly comparable to the fine structure splitting in the $2p$ states (0.34 $\mathrm{cm}^{-1}$), and therefore both the $2p\ ^2\mathrm{P}_{1/2}$ and $2p\ ^2\mathrm{P}_{3/2}$ states are populated. The excitation laser beam has a spot diameter $\sim$1 mm and its energy is adjusted such that the excitation signal from the $2p\ ^2\mathrm{P}_{1/2,3/2}$ states can be easily saturated. This state is subsequently ionized by a second laser of variable frequency and pulse energy. Both the laser beams are linearly polarized, as the polarization vectors affect the measurement of cross-section. The area of the overlap region in the confocal limit is calculated using the following relation [25,26]

$$A = \pi\omega_0^2 \left[1 + \left(\frac{\lambda Z}{\pi\omega_0^2}\right)\right]^2. \quad (3)$$

Here $\omega_0 = f\lambda/\pi\omega_s$ is the beam waist, where $\omega_s$ is the spot size of the beam on the focusing lens. The cross-sectional area corresponding to 349.8 nm is calculated as 0.0032(1) $\mathrm{cm}^2$ using the spot size 0.0174 cm at a focal length of 50 cm and $Z$ is taken as the effective length of the heating zone. The energy of the ionizing laser beam is varied by inserting the neutral density filters (Edmond Optics) and measured using an energy meter (R-752, Universal Radiometer) within $\pm 5\%$ variation. We registered the signal intensities against the pulse energies of the ionizing laser. The atomic photoionization cross-section depends upon the excess photoionization energy from the ionization limit. By changing the photon energy, the frequency dependence of the cross-section can be studied. A smooth frequency dependence has been observed because all the alkali atoms possess a single valance electron [5]. For atoms with several valence electrons the pattern of the photoionization spectra changes considerably due to the presence of auto-ionizing resonances either due to double excitation or inner shell excitation. We have determined the absolute values of the photoionization cross-sections at three different ionizing laser wavelengths: 349.8 nm, 335 nm and 266 nm. The corresponding graphs to achieve saturation at these wavelengths are shown in Figures 2a–2c. From these figures it is evident that as the energy of the ionizing laser increases, the ion signal increases up to a certain value and then the signal stops increasing further. At this point the photoionization from the excited state reaches its maximum and saturation sets in. The threshold laser energy of the ionizing laser at which the saturation just occurs is relatively lower at 349.8 nm, and a higher energy is required at 266 nm to ionize the excited atoms. The difference in the threshold energy is attributed to the corresponding photoionization cross-sections. The solid line in Figure 2, which passes through the data points, is the least squares fit to equation (1). The corresponding photoionization cross-sections at the ionizing wavelengths are thus 14.8 Mb, 12.7 Mb and 8.4 Mb respectively.

A comparison of the available experimental and theoretical data is presented in Figure 3. The continuous curve is taken from the theoretical work of Lahiri and Manson [14], which covers the energy from the first ionization threshold up to 0.3 Ry, whereas the experimental data points are confined to a small region above the threshold. Karlov et al. [9] measured the photoionization cross-section from the $2p$ excited state at the ionizing wavelength 337.1 nm as $10 \pm 3$ Mb which is about 4 Mb lower than the theoretical results. Similarly, Wippel et al. [17] reported the photoionization cross-section from the $2p$ excited state at the ionizing wave length 334.47 nm as 16.2 Mb and at 335.85 nm as 18.3 Mb using the magneto optical trap (MOT) method. These values of the cross-sections are close to the experimental value reported

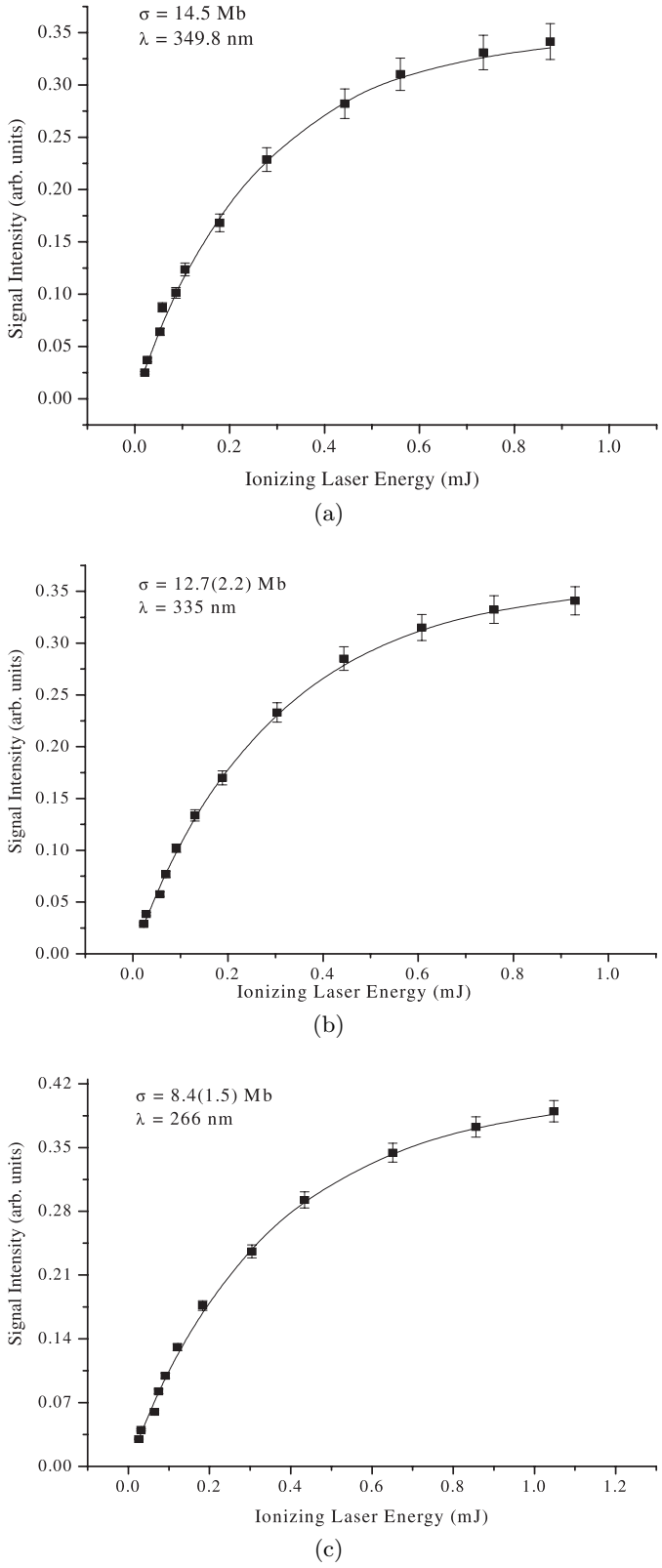


**Fig. 2.** The photoionization data for the $2p\ ^2P_{1/2,3/2}$ state of lithium atoms corresponding to $2s\ ^2S_{1/2} \rightarrow 2p\ ^2P_{1/2,3/2}$ transitions at ionizing lasers (a) 349.8 nm (b) 335 nm and (c) 266 nm. The solid line is the least squares fit to equation (1) to the observed data for extracting the photoionization cross-section.

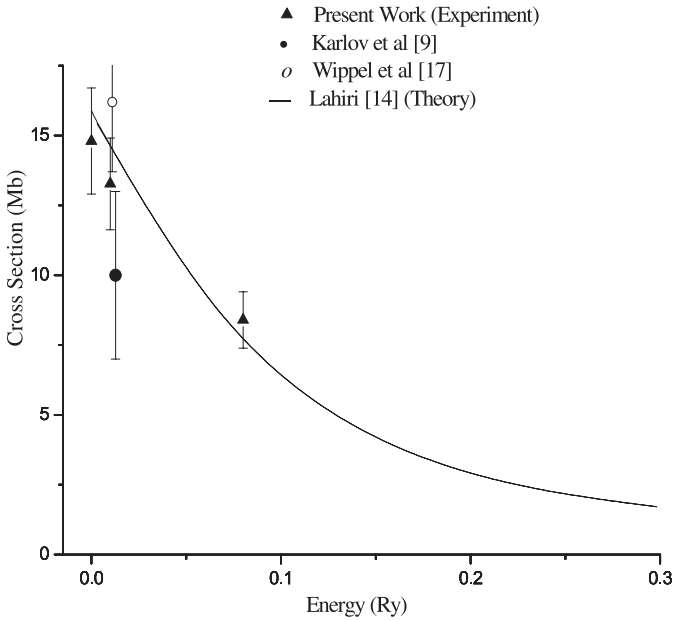


**Fig. 3.** A comparison of the photoionization cross-sections of the present and previous work for the $2p$ excited state of lithium. The (▲) is the present work, (● and ○) are the experimental results with respective error bars taken from references [9,17] respectively. The solid curve is the calculated result presented by Lahiri and Manson [14].

by Rothe [4] and the theoretical value reported by Lahiri and Manson [14]. The measured values of photoionization cross-sections in the present work are very close to the theoretical curve presented by Lahiri and Manson [14]. Despite some limitations in the measurement of the photoionization cross-section by the saturation technique, our measured values are reasonably in agreement with the experimental and theoretical values.

In the second set of experiments we measured the photoionization cross-sections from the $3d\ ^2D_{3/2,5/2}$ state as a function of the laser frequency above the first ionization threshold. The energy level diagram showing the relevant transitions is shown in Figure 4. In a two-photon excitation process, we have excited the atoms from the ground state to the even parity $3d\ ^2D_{3/2,5/2}$ states at 31283.08 $cm^{-1}$ and 31283.12 $cm^{-1}$ respectively [28]. As the fine structure splitting in the $3d$ states is 0.04 $cm^{-1}$, it could not be resolved in the present work. Therefore, both the $3d\ ^2D_{3/2}$ and $3d\ ^2D_{5/2}$ states are populated. According to the two-photon excitation selection rules, $\Delta\ell = 0, \pm 2$, $\Delta J = 0, \pm 2$, $\Delta L = 0, \pm 2$, $\Delta S = 0$ and odd $\leftrightarrow$ odd or even $\leftrightarrow$ even parity, only the $3d\ ^2D_{5/2}$ state can be accessed from the ground state. In the present work we have determined the absolute values of the photoionization cross-section from the $3d\ ^2D_{5/2}$ state at different ionizing laser wavelengths. A typical data for the photoionization of $3d$ at ionizing laser wavelength 780 nm is shown in Figure 5. The solid line, which passes through the data points, is the least squares fit to equation (1). The cross-sectional area is calculated as 0.0036 (1) $cm^2$

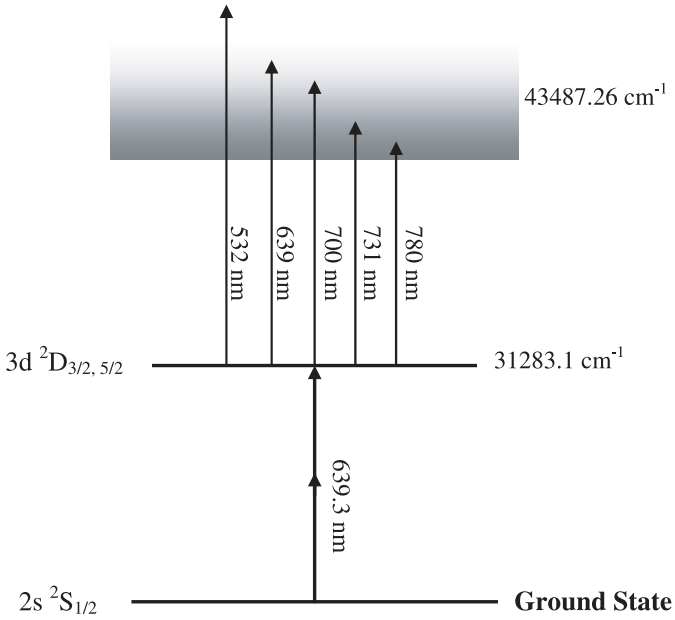


**Fig. 4.** Energy level diagram for the measurement of the photoionization cross-section from the $3d$ excited state.

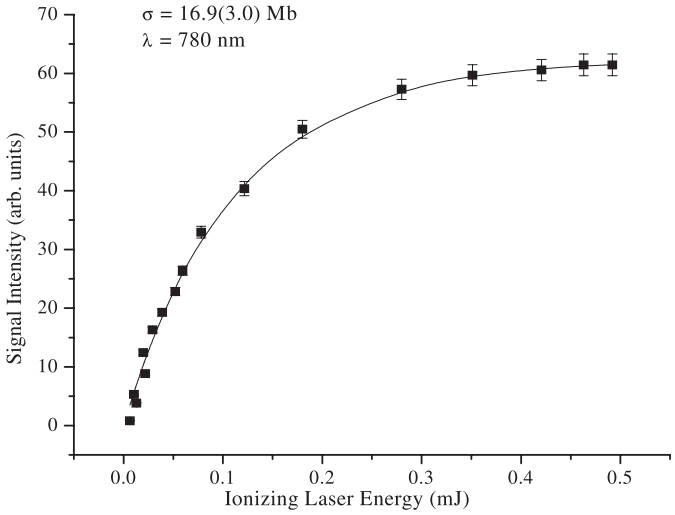


**Fig. 5.** The photoionization data for the state $3d\ ^2D_{3/2,5/2}$ of lithium atoms corresponding to the $2s\ ^2S_{1/2} \xrightarrow{2\hbar\omega} 3d\ ^2D_{5/2}$ transition at 780 nm ionizing laser. The solid line is the least squares fit to equation (1) to the observed data for extracting the photoionization cross-section.

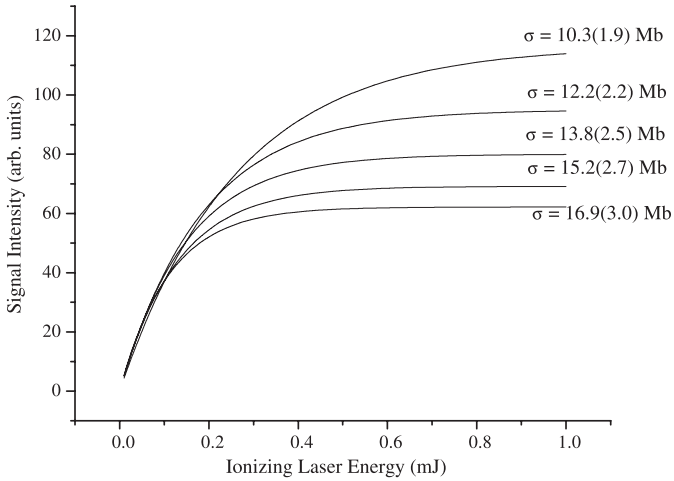


**Fig. 6.** A set of photoionization signals for the $3d\ ^2D_{5/2}$ states of lithium as a function of ionizing lasers energies. The top signal is at 532 nm, while the signal at the bottom is at 780 nm.

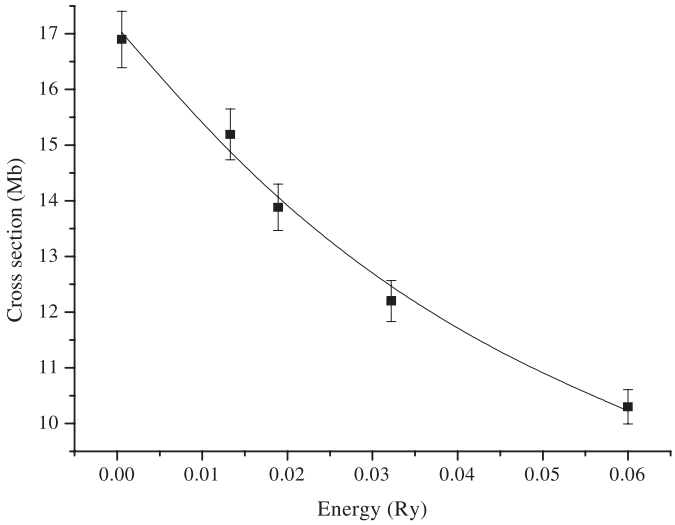


**Fig. 7.** A plot of the photoionization cross-section of the $3d\ ^2D_{3/2,5/2}$ states of lithium versus the energy in Rydberg above the ionization threshold. The solid line that passes through the data points represents the first order exponential decay law.

by using spot size 0.0374 cm at a focal length of 50 cm for ionizing wavelength of 780 nm. Fitting of the experimental data for the $3d$ state yields the photoionization cross-section $\sigma = 16.9$ Mb. Similarly we registered the saturated ionization signals at 731 nm, 700 nm, 639 nm and 532 nm and the corresponding cross-sections at these wavelengths are 15.2 Mb, 13.8 Mb, 12.2 Mb and 10.3 Mb respectively. The fitted curves to the experimental data at five ionizing laser wavelengths, as mentioned above are shown in Figure 6. It is evident from the figure that as the laser intensity increases, the ion signal increases up to a certain value very swiftly and then changes slowly. The multi-step photoionization depends upon the flux of the ionizing laser pulse, which enables absolute measurement of the photoionization cross-section. At saturation ionization the photon flux of the ionizing laser equalizes the population of the intermediate states. In the present case full saturation could not be obtained due to insufficient dye laser energy and Gaussian wing of laser pulse distribution. The measured photoionization cross-section as a function of excess energy above the first ionization threshold up to 0.06 Rydberg is shown in Figure 7. The solid line which passes through the experimental data points is

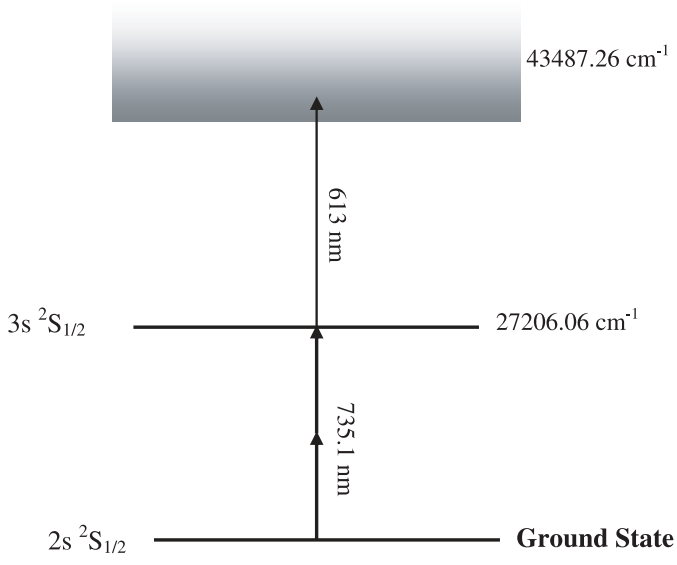


**Fig. 8.** Energy level diagram for the measurement of the photoionization cross-section from the $3s\ ^2S_{1/2}$ states by two-photon excitation from the ground state.

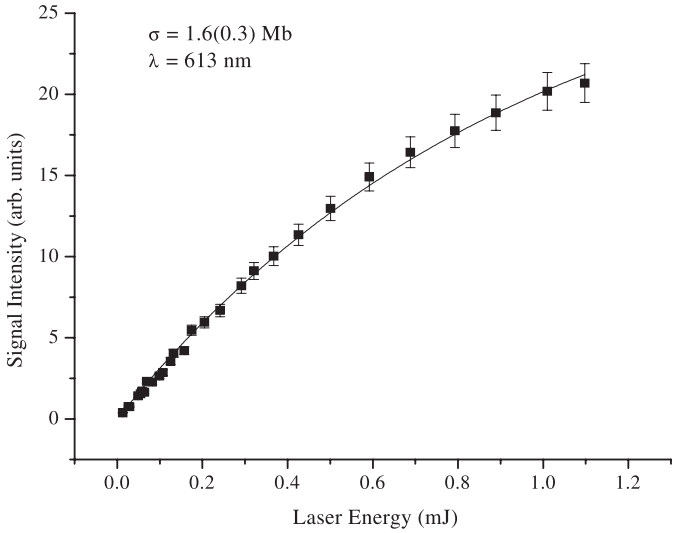


**Fig. 9.** The photoionization data for the $3s\ ^2S_{1/2}$ state of lithium atoms corresponding to the $2s\ ^2S_{1/2} \xrightarrow{2\hbar\omega} 3s\ ^2S_{1/2}$ transition at 613 nm ionizing laser.

the fit of an exponential law, demonstrating that the photoionization cross-section above the ionization threshold changes very smoothly. Very little information is available about the cross-sections from the $3d$ excited states of lithium. Aymar et al. [7] calculated the photoionization cross-section of $3d$ at ionization threshold as 18.2 Mb whereas Gezalov and Ivanova [29] reported the threshold to be 17.5 Mb. The cross-section determined in the present work near the ionization threshold is 16.9 Mb which is in good agreement with those obtained in the theoretical work [7,29]. Since the photoionization cross-sections from the $3d$ excited state above the threshold at different wave lengths are explored for the first time, a comparison is not possible. It will be interesting to have some theoretical work to compare with the experimental data.

Similarly, we measured the photoionization cross-sections from the $3s\ ^2S_{1/2}$ excited state of lithium by using two-photon excitation from the ground state. The $3s\ ^2S_{1/2}$ state is populated using two-photon excitation from the ground state by tuning the dye laser at wavelength 735.1 nm. The second dye laser is tuned to achieve the ionization from the $3s\ ^2S_{1/2}$ excited states at 613 nm. We recorded the signal intensity versus the pulse energy of the ionizing laser beam by inserting neutral density filters in the optical path of the ionizing laser. The cross-sectional area of ionizing laser (613 nm) is calculated by using equation (3), as 0.0023(1) $cm^2$. The beam waist ($\omega_0$) is 0.0261 for the spot size 0.0374 cm. The ion signal is registered on a storage oscilloscope and on a computer for further processing. The measured data points, ionization signal verses the ionizing laser energy is shown in Figure 9. The solid line that passes through the experimental data points is the least square fit to equation (1). This fitting of the experimental data gives the photoionization cross-section $\sigma = 1.6$ Mb. For the $3s$ excited state of lithium, no previous experimental results are available but theoretical results at the first ionization threshold were reported by many groups. Our measured value of the photoionization cross-section at the ionization threshold is in good agreement, within the experimental uncertainty, to those reported in the theoretical work ([7,14] and references therein).

The experimental and theoretical values for photoionization cross-sections at different ionizing laser wavelengths are presented in Table 1. The overall uncertainty in all the results is about 18%, which is attributed to errors in the measurement of the cross-sectional area of the laser beam (10%), the laser energy (5%) and the calibration of the detection system (2%). In the measurements of the photoionization cross-sections the role of recombination processes is very important near the ionization threshold. The cross-section for radiative recombination can be extracted from the detailed balance principle as described by Lahiri and Manson [7]. In the low energy region radiative recombination dominates the recombination processes. The photoionization cross-section has been measured for $3s$ ($\ell = 0$), $2p$ ($\ell = 1$) and $3d$($\ell = 2$) near the threshold as 1.6 Mb, 14.8 Mb and 16.9 Mb respectively. Consequently, the radiative recombination cross-section is smaller for $3s$ and dominating for the $3d$ state, in accordance with the theoretical calculations [7].

In conclusion we have been able to extract the values of the photoionization cross-sections from the $2p\ ^2P_{1/2,3/2}$ and $3d\ ^2D_{3/2,5/2}$ excited states of lithium as a function of the ionizing laser frequency above the ionization threshold. The measured values are in good agreement with that reported in the literature, where available. The photoionization cross-section from the $3s\ ^2S_{1/2}$ excited state is estimated at 613 nm as 1.6 Mb at threshold.

**Table 1.** Photoionization cross-sections from the excited states in lithium.

| | Present work | | Previous work | |
|---|---|---|---|---|
| state | wavelength (nm) | cross-section (Mb) | wavelength (nm) | cross-section (Mb) |
| $2p\ ^2P_{1/2,3/2}$ | 266.0 | 8.4 (1.5) | 334.47 | 16.2 [17] Exp |
| | 335.0 | 12.7 (2.2) | 337.10 | 10.0 [9] Exp |
| | | | 349.8 | 19.7 [4] Exp |
| | 349.8 | 14.8 (2.6) | 349.8 | 15.2 [7] (Th.) |
| | | | | 15.2 [27] (Th.) |
| $3d\ ^2D_{3/2,5/2}$ | 532 | 10.3 (1.9) | 819.4 | 18.2 [7] (Th.) |
| | 639 | 12.2 (2.2) | 819.4 | 17.5 [29] (Th.) |
| | 700 | 13.8 (2.5) | | |
| | 731 | 15.2 (2.7) | | |
| | 780 | 16.9 (3.0) | | |
| $3s\ ^2S_{1/2}$ | 613 | 1.6 (0.3) | 614.2 | 1.27 [30] (Th.) |
| | | | | 1.42 [31,32] (Th.) |
| | | | | 1.17 [29] (Th.) |
| | | | | 1.48 [7] (Th.) |

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