Photoionization cross section measurements of the $3p\ ^{1,3}P$ excited states of helium in the near-threshold region

Shahid Hussain, M. Saleem, M. Rafiq, and M. A. Baig*
Atomic and Molecular Physics Laboratory, Department of Physics, Quaid-i-Azam University, Islamabad 45320, Pakistan
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We present measurements of photoionization cross sections of the $3p\ ^1P$ and $3p\ ^3P$ excited states of helium, at threshold and near-threshold region (0–0.2 Ry). The experiments have been performed using a dc glow discharge and employed the saturation technique to determine the photoionization cross sections. A smooth frequency dependence of the cross section has been observed for both the excited states in accordance to the theoretical calculations. The measured values of the photoionization cross section, using a simple experimental setup, are in good agreement with the earlier reported theoretical and experimental values.

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INTRODUCTION

Helium possessing filled s subshell in the ground state is the simplest element after hydrogen. It is an ideal system for the study of atom-light interaction and in particular for the photoionization processes [1]. Photoionization from the excited states of helium plays an important role in the radiation transfer through laboratory plasmas and hot stellar atmosphere [2]. Accurate values of the photoionization cross sections of the helium excited states is needed for a quantitative understanding of the laser induced processes in a low-pressure helium plasma [3]. The photoionization cross section of the ground state of helium has been extensively studied [4]. The knowledge of the photoionization cross section of the excited states of helium has progressed mostly by theoretical efforts [5–12] and only a few experimental studies have been devoted to the excited states of helium [13–15]. The absolute cross sections for photoionization of the $2s\ ^1S_0$ and $^3S_1$ metastable states, from threshold to 240 nm were reported by Stebbings *et al.* [13] whereas that of the $3p\ ^{1,3}P$, $4p\ ^{1,3}P$, and $5p\ ^{1,3}P$ states were measured by Dunning and Stebbings [14]. The photoionization cross sections for each of these states were determined at the wavelength used for its excitation from the $2\ ^{1,3}S$ metastable state. Gisselbrecht *et al.* [15] experimentally determined the absolute photoionization cross sections of the $1s2p\ ^1P$ and $1s3p\ ^1P$ excited states in the region close to the threshold (from 0 to 2 eV). The intermediate excited states were achieved by the photoabsorption of a high-order harmonic of an intense picosecond tunable laser and subsequently ionized by absorption of photons of several fixed frequencies, ranging from the near infrared to ultraviolet.

The excited states of inert gases lie in the extreme ultraviolet (xuv), which are difficult to optically excite by absorbing a single photon due to the nonavailability of the tunable lasers in the xuv region. One of the techniques to achieve coherent and linearly polarized xuv light is based on the generation of high-order harmonics of an intense laser pulse. This technique has been used for the spectroscopic studies of the rare gas atoms [16]. Interestingly, the $2s\ ^{1,3}S$ metastable levels of helium gets significantly populated in a low-pressure glow discharge and from these levels the highly excited states can be easily accessed using the laser optogalvanic effect [17–19]. Babin and Gagné [20] used a hollow cathode discharge cell for the measurement of the photoionization cross sections of refractory elements and Stockhausen *et al.* [21] used a hollow cathode lamp to measure the photoionization cross section of the autoionizing lines of copper. The optogalvanic method, in conjunction with hollow cathode discharges has also been used for the study of the photoionization processes in complex atoms [22].

In the present work we have experimentally determined the photoionization cross sections of the $3p\ ^{1,3}P$ excited states of helium at several laser frequencies from threshold up to 0.2 Ry. The $3p\ ^{1,3}P$ states are populated by absorbing a single photon at 501.7 nm and 388.9 nm via the $2s\ ^{1,3}S$ metastable states, respectively. These levels are subsequently ionized by a probe laser with variable energy densities and frequencies. We have found a smooth wavelength dependence of the photoionization cross sections for both the excited states. The measured values of the photoionization cross are then compared with the earlier available experimental and theoretical work.

EXPERIMENTAL DETAILS

A schematic diagram of the experimental setup is shown in Fig. 1. It includes two different optical paths starting from the same primary Nd:YAG laser (Quanta Ray GCR-11) operating at 10 Hz repetition rate and 5 ns pulse duration. The laser system is equipped with potassium dihydrogen phosphate (KDP) (kalium-dihydrogen phosphat) type-II crystals for SHG (second harmonic generation) and THG (third harmonic generation) and a wavelength separation assembly. The SHG at 532 nm and THG at 355 nm were used to pump two locally made Hanna type [23,24] dye lasers equipped with holographic gratings and prism beam expanders. The linewidth of the dye laser was $\leqslant 0.3\ \mathrm{cm}^{-1}$. The dyes LD-820, LD-390, DCM, Coumarin 500 and Pyridine-2 dissolved in methanol were used to acquire the required ionizing laser

*Corresponding author. FAX: +92 51 9210256. Email address: baig@qau.edu.pk

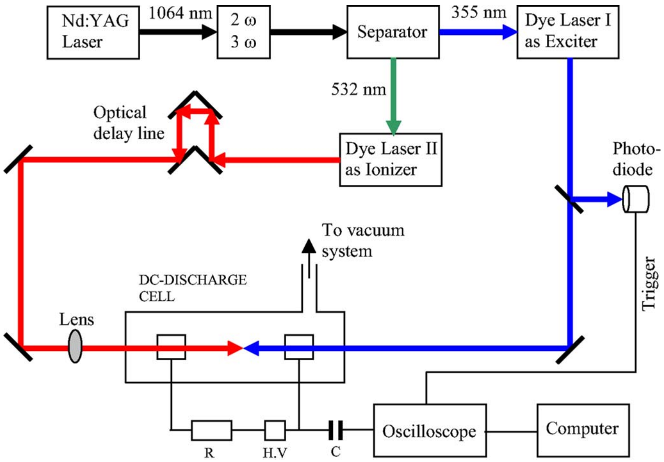

FIG. 1. (Color online) Schematic diagram of the experimental setup for two-step photoionization of helium.

wavelengths. The frequency doubling of the dye laser, in some cases, was achieved by a BBO type-I crystal. The dye laser wavelength was determined by a spectrometer (Ocean Optics, HR2000) equipped with a 600 lines/mm grating. The variation in the amplitude of the optogalvanic signals with the laser intensity was recorded using a digital storage oscilloscope (TDS 2024) and a computer through RS232 interface. The intensity of the ionizing laser was varied by inserting the neutral density filters (Edmund Optics) and on each insertion the energy was measured by an energy meter (R-752, Universal Radiometer).

A homemade dc discharge cell, with quartz windows at its two ends, was used in the present experiments. It consisted of a Pyrex tube, 15 cm long and 2.5 cm in diameter, having two hollow nickel foil cylindrical electrodes, 10 mm in diameter and 10 mm long, and spaced by ≈25 mm. Typical discharge conditions were 360 V, 2 mA current, and ≈3 mb helium gas pressure. The discharge conditions were optimized so that a cylindrical shaped negative glow formed at the electrodes axes and filled the space between the electrodes.

In order to excite the $3p\ ^1P$ state at 501.7 nm one of the dye lasers was charged with Coumarin 500 dissolved in methanol and pumped with the third harmonic (355 nm) of the Nd:YAG laser. The second harmonic of the Nd:YAG laser at 532 nm was used to pump a second Hanna type [24] dye laser. The output from this dye laser was used for the ionization step. In some experiments, the SHG at 532 nm and the THG at 355 nm of the Nd:YAG laser, were also used in the ionization step.

The two laser beams (the exciter and ionizer), entering the discharge cell from the opposite sides, overlap at the center of the negative glow. The relative delay between the exciter and ionizer laser pulses was controlled and varied by an optical delay line. The temporal overlap of both the laser pulses was checked using a photodiode.

RESULTS AND DISCUSSION

The two-step photoionization scheme is shown in Fig. 2.

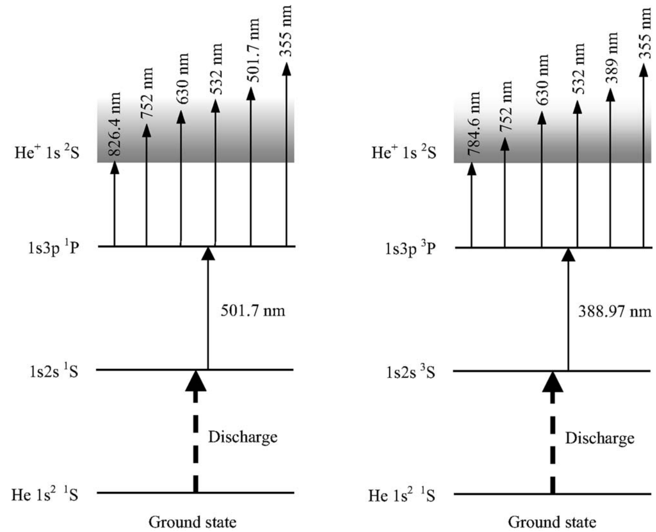

FIG. 2. Energy level diagram for the measurement of photoionization cross section from $3p\ ^{1,3}P$ excited states of helium.

In the first step, the helium atoms are optically excited from the collisionally populated $1s2s\ ^{1,3}S$ metastable states to the $1s3p\ ^{1,3}P$ states. In the second step, the excited electrons are promoted to the $He^+\ 1s\ ^2S$ continuum by a second laser pulse. By varying the frequency (wavelength) of the ionizing laser the electrons of different kinetic energies are produced and different regions of the continuum are investigated. Since only the $He^+\ 1s\ ^2S$ continuum can be reached with the available energies of the ionizing laser used in the present studies, the number of electrons produced and hence the optogalvanic signal directly reflects the photoionization cross section in the investigated regions.

Our determination of the absolute photoionization cross section is based on the saturation method [25–28]. Burkhardt *et al.* [25] applied the two-step ionization technique to measure the absolute cross section from the excited states of sodium, potassium, and barium. In these experiments it was assumed that the intensity of the ionizing laser is higher than required for saturating the resonance transitions, 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

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

Here e (Coulomb) is the electronic charge, N_0 (cm^{-3}) is the density of the excited atoms, A (cm^2) is the cross sectional area of the ionizing laser beam, U (joule) is the total energy per ionizing laser pulse, $\hbar\omega$ (joule) is the energy per photon of the ionizing laser beam, V_{vol} (cm^3) is the laser interaction volume, and σ (cm^2) is the absolute cross section for photoionization. The major advantage of this technique is that an absolute measurement of the cross section can be made even if the measured quantity, which is proportional to Q in Eq. (1), is arbitrary.

He *et al.* [26] measured the absolute photoionization cross section of the $6s6p\ ^1P_1$ excited state of barium using

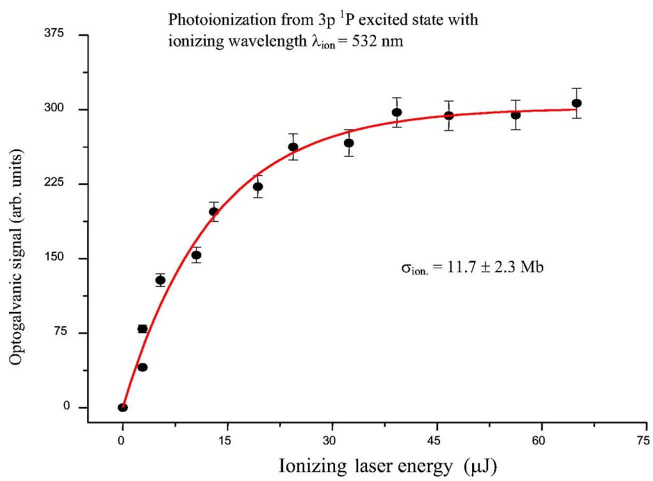

FIG. 3. (Color online) The photoionization data for the $3p\ ^1P$ excited state of helium with the ionizing laser set to a wavelength of 532 nm. The solid line is the least squares fit to Eq. (1) to the observed data for extracting the photoionization cross section. The error limits on the data results from pulse-to-pulse fluctuations in the signal.

the same technique [25] but accounting for the effects for the Gaussian laser intensity distribution. An expression similar to that of Burkhardt *et al.* [25] was used by Xu *et al.* [29] to measure the photoionization cross sections of the autoionizing states of lutetium. Recently, we have exploited this technique to measure the photoionization cross section and the optical oscillator strength of the autoionizing resonances in neon using a hollow cathode dc discharge [30]. In the present experiment we have determined the photoionization cross section using Eq. (1) under the best alignment conditions for the Gaussian laser intensity distribution. This requires accurate measurements of the ionizing laser energy as well as characterization of the spatial profiles of both the exciter and ionizer laser pulses in the interaction region. The uncertainty in the energy determination is mostly owing to the energy fluctuations in the fundamental Nd:YAG laser and in the measuring instruments. A lens of focal length 50 cm was used in the ionization step to meet the power requirements for saturation. The spot size of the exciter laser is ≈3 mm, which is much larger than the spot size of the ionizing laser. This eliminates the problems associated with the spatial overlapping of the exciter and the ionizer laser pulses. The area of the overlap region in the confocal limit is calculated by using the relation [31]

$$A = \pi\omega_0{}^2\left[1+\left(\frac{\lambda_{io}z}{\pi\omega_0{}^2}\right)^2\right]. \qquad (2)$$

Here z is the distance on the beam propagation axis from the focus, $\omega_0 = f\lambda_{io}/\pi\omega_s$ is the beam waist at $z=0$, ω_s is the spot size of the ionizing laser beam on the focusing lens, f is the focal length, and λ_{io} is the wavelength of the ionizing laser.

The amplitude of the optogalvanic signals is plotted as a function of energy of the ionizing laser. A typical experimental curve for the $3p\ ^1P$ excited states of helium using the ionizing laser wavelength 532 nm at different energy range

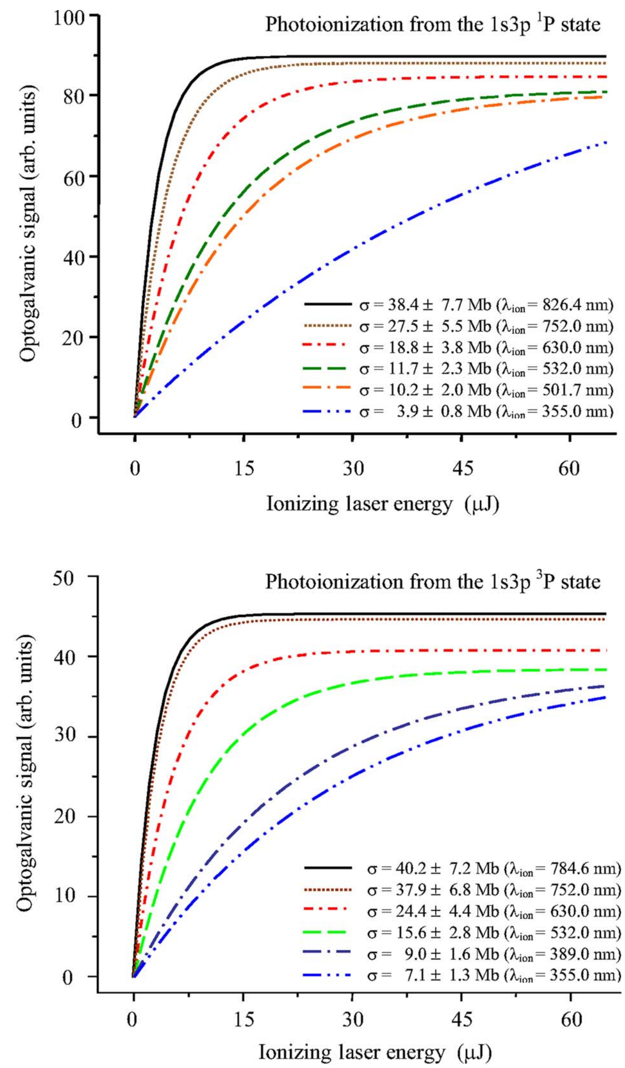

FIG. 4. (Color online) (a) The fitted curves to the experimental data of the optogalvanic signals against the energy of the ionizing laser for six different wavelengths at 826.358, 752, 630, 532, 501.7, and 355 nm using Eq. (1). These fitted curves are used to extract the photoionization cross section of the $3p\ ^1P$ excited state of helium. (b) The fitted curves to the experimental data of the optogalvanic signals against the energy of the ionizing laser for six different wavelengths at 784.55, 752, 630, 532, 389, and 355 nm using Eq. (1). These fitted curves are used to extract the photoionization cross section of the $3p\ ^3P$ excited state of helium.

up to 75 μJ is presented in Fig. 3. The solid line that passes through the experimental data points is the least square fit to Eq. (1). It is evident that the optogalvanic signal first increases linearly with the intensity of the ionizing laser and then saturates, i.e., the optogalvanic signal stops to increase by further increase in the ionizing laser intensity. The optogalvanic signal due to photoionization from the $3p\ ^1P_1$ excited state at six ionizing laser wavelengths 826.4, 752, 630, 532, 501.7, and 355 nm are shown in Fig. 4(a). The corresponding optogalvanic signal due to photoionization from the $3p\ ^3P_1$ excited state at 784.6, 752, 630, 532, 389, and 355 nm are shown in Fig. 4(b). It is evident that the photoion current increases with an increase in the energy of the ionizer

TABLE I. Experimental data for absolute photoionization cross section from excited states of helium.

	Present work		Previous work
State	Ionizing wavelength (nm)	Photoionization cross section (Mb)	Photoionization cross section (Mb)
$3p\ ^1P$	826.4	38.4±7.7	
	752.0	27.5±5.5	2.4 [15]
	630.0	18.8±3.8	
	532.0	11.7±2.3	10.5 [15]
	501.7	10.2±2.0	10±2 [14]
	355.0	3.9±0.8	4.2 [15]
$3p\ ^3P$	784.6	40.2±7.2	
	752.0	37.9±6.8	
	630.0	24.4±4.4	
	532.0	15.6±2.8	
	389.0	9.0±1.6	8.7±2 [14]
	355.0	7.1±1.3	

laser until it approaches saturation. Each curve is a least square fit of Eq. (1) to the experimental data points acquired at a particular ionizer laser wavelength at different energies. The photoionization cross sections determined from the fitting procedure, along with the earlier reported values, are summarized in Table I.

Since the He* $1s3p\ ^3P$ has longer lifetime (1.05 μs) than the exciter laser pulse duration, therefore the spontaneous emission can be nullified. However, the spontaneous emission from the He* $1s3p\ ^1P$ cannot be ignored because of its shorter lifetime (1.7 ns). The loss of the excited atoms due to spontaneous emission is compensated by the excitation of further $3p\ ^1P$ state from the metastable state in order to maintain saturation of the transition [14].

The maximum overall uncertainty in the determination of the absolute cross section is estimated to be 18% [32] for the ^3P excited state, which is attributed to the experimental errors in the measurements of the laser energy, the cross sectional area of the ionizing laser beam in the interaction region and the collisional losses. Whereas, for the $3p\ ^1P_1$ excited state, the uncertainty in the measurement of cross section is estimated as 20%, which includes an additional error due to the spontaneous emission during the laser pulse [14].

The photoionization cross sections for the excited states have also attracted considerable attention based on configuration interactions [11,12] and multiconfiguration Hartree-Fock method [33]. Most of the calculations addressed the total cross sections considering the interaction/ionization by an unpolarized radiation. These include all the allowed transitions between the magnetic sublevels following $\Delta m=0,\pm 1$ selection rules. In the present experiment we have used two dye lasers, both are linearly polarized and have parallel polarization vectors. Under these conditions only transitions between the magnetic sublevels $\Delta m=0$ are allowed. The helium $3p\ ^{1,3}P$ can be ionized towards two

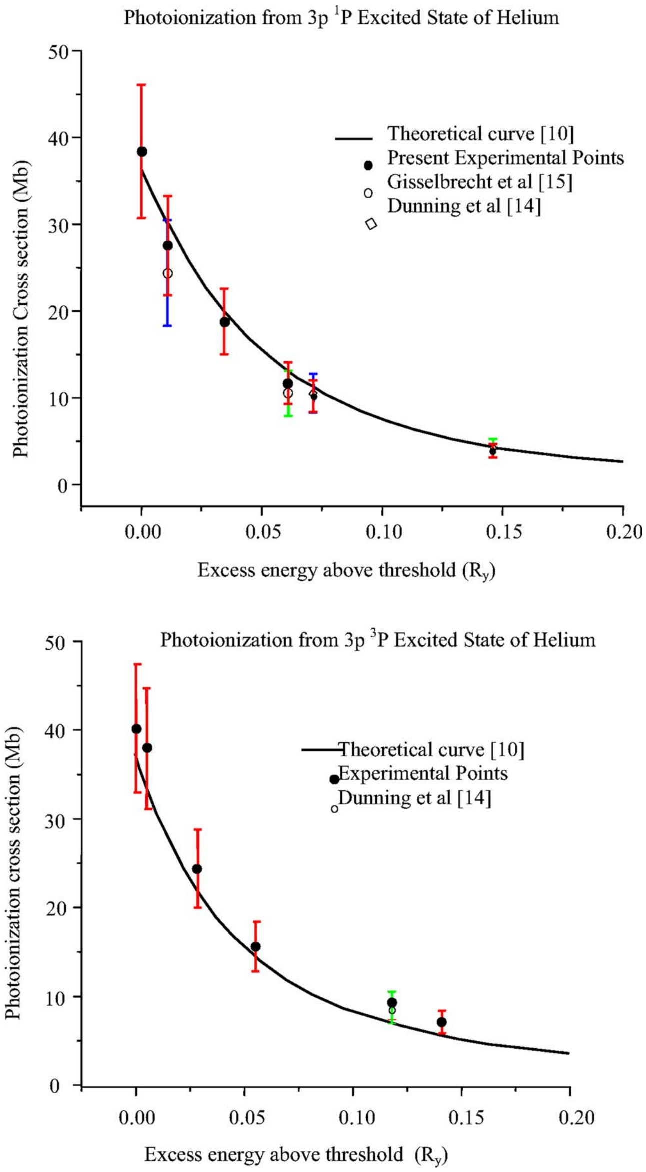

FIG. 5. (Color online) Comparison of the present and previous experimentally measured values of the absolute photoionization cross section from $3p\ ^{1,3}P$ with the theoretical curve of Jacobs [10] against the excess photon energy above the first ionization threshold.

ionization continua corresponding to outgoing εs and εd waves. The partial photoionization cross sections for the $np\ ^{1,3}P$ excited states of helium have been calculated for parallel polarization by Jacobs [10]. It was inferred that the contributions of S and D waves might be combined to obtain the total cross section for arbitrary polarization states of the excited helium atoms and of the ionizing radiation.

The photoionization cross section from the $3p\ ^1P$ excited state of helium measured at six wavelengths (826.4, 752, 630, 532, 501.7, and 355 nm) of the ionizing laser which covers energy from the first ionization threshold up to 0.2 Ry excess energy is shown in Fig. 5(a). The cross section varies with the ionizing laser wavelength and display an overall decrease from a maximum value ≈40 Mb (1 Mb=10^{-18} cm^2) near threshold to ≈4 Mb at 0.15 Ry above the ionization threshold. The continuous line is the theoretical calculations [10]. Gisselbrecht *et al.* [15] reported the $3p\ ^1P$ cross section at three ionizing wavelengths 752, 532, and 355 nm. Our measured values at 532 and 355 nm are in excellent agreement with those reported by Gisselbrecht *et al.* [15]. However, the measured value at 752 nm is slightly higher but is within our experimental error. Dunning and Stebbings [14] reported the value of $3p\ ^1P$ excited state at only one wavelength 501.7 nm, i.e., at the same wavelength used to excite the $3p\ ^1P$ state. Our experimentally measured value at 501.7 nm is also in excellent agreement with that by Dunning and Stebbings [14].

The photoionization cross section from the $3p\ ^3P$ excited state of helium measured at six wavelengths (784.6, 752, 630, 532, 389, and 355 nm) of the ionizing laser which covers energy from the first ionization threshold up to 0.2 Ry excess energy is shown in Fig. 5(b). The cross section from $3p\ ^3P$ at threshold is nearly the same ($\approx$40 Mb) as from $3p\ ^1P$ and decreases smoothly with the ionizing laser wavelength above the ionization threshold. The solid line is the theoretical curve by Jacobs [10]. There exists only one measurement of the cross section of the $3p\ ^3P$ excited state and also only at one ionizing laser wavelength (389 nm) [14]. The value presently reported at 389 nm is in excellent agreement to that of reported by Dunning and Stebbings [14]. However, the measurements (present work) extend the values of cross section from threshold to 0.2 Ry at six different ionizing laser wavelengths and exhibit excellent agreement with the theoretical work [10].

In conclusion, we have used a simple experimental arrangement (dc glow discharge) and determined the absolute photoionization cross sections from the $3p\ ^{1,3}P$ excited states of helium which are in good agreement with the earlier experimental and theoretical studies. The availability of metastable atoms in a glow discharge makes possible to access the highly excited states of rare gases with tunable uv lasers. It is therefore, an alternate simple technique to approach the highly excited states instead of generating higher order harmonics that requires a lot of sophisticated experimentation. This technique can be extended to investigate the photoionization cross sections of other rare gases. The work on the determination of absolute oscillator strength of the helium Rydberg series is in progress.

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