Evidence of a Cooper minimum in the photoionization from the $7s\ ^2S_{1/2}$ excited state of potassium

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The existence of a Cooper minimum in the experimentally measured photoionization cross sections for the $7s\ ^2S_{1/2}$ excited state of potassium has been reported using a two-step laser excitation technique in conjunction with a time-of-flight mass spectrometer. The photoionization cross sections for the $7s\ ^2S_{1/2}$ state have been determined as $(9.78 \pm 0.01) \times 10^{-2}$, $(3.30 \pm 0.49) \times 10^{-4}$, $(5.47 \pm 0.82) \times 10^{-3}$, and $(2.06 \pm 0.30) \times 10^{-2}$ Mb at the ionizing laser wavelengths 1064, 700, 532, and 355 nm, respectively, that validate the presence of a Cooper minimum at $\cong$1.25 eV above the first ionization threshold. The number density has been determined as $N_0 = 2 \times 10^{11}$ cm^{-3}. The experimentally determined absolute values of the cross sections are in qualitative agreement with the theoretical predictions.

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I. INTRODUCTION

The manifestation of a Cooper minimum in photoionization studies and the transition probabilities from the ground states of alkali metals has been a great interest of researchers for some time. However, in recent years the Cooper minimum has attracted a lot of attention due to its existence in the transitions from the excited state of alkali metals. The Cooper minimum is a dynamical phenomenon that can be encountered in the photoionization process which provides valuable information about the electron correlation and relativistic effects. These minima arise when the matrix element of the photoionization process vanishes as a result of positive and negative contributions to their radial parts caused by the overlap between the discrete and the continuum wave functions. The existence of the Cooper minimum in photoionization cross sections has been investigated experimentally and theoretically [1–11] in transitions from the ground state of alkali metals and in particular for potassium atoms. However, the Cooper minimum in the transitions from the excited states of alkali metals has been reported [12–14] recently; these reports presented experimental evidence for verification of this phenomenon.

Historically, the presence of a minimum was first observed by Ditchburn *et al*. [1] in their measurements of the continuous absorption of light in potassium vapors, extending into the vacuum ultraviolet region. They found that the absorption coefficient of the potassium vapors decreases to a minimum value and then increases rapidly with the decreasing wavelength. Bates [2] explained the experimentally observed minima by the theoretical calculations of the photoionization of atomic potassium using the Quantal Methods. Subsequently, these observed minima were explained by Cooper [3] and Fano and Cooper [4] in their detailed theoretical investigations and named the Cooper minimum (CM). Hudson and Carter [5] reported the photoionization cross sections for the sodium ground state showing the Cooper minimum that was in good agreement with the theoretical calculations of Aymar *et al*. [6], confirming the accuracy of the theoretical model. Baum *et al*. [7] investigated the spin-orbit interaction for the *p*-state continuum of heavy alkali metals K, Rb, and Cs in a photoionization experiment using spin-polarized alkali-metal atoms and circularly polarized light. They used the knowledge of the spin-orbit perturbation parameter as a function of energy to establish the accurate position of the Cooper minimum and to estimate the magnitude of the cross section at the minimum. Heinzmann *et al*. [8] predicted that the Cooper minima of the photoionization cross section should also exist for the l→(l–1) transitions. The theoretical conclusions were supported by the measurements of the spin polarization of photoelectrons that had been ejected from the thallium atoms by the circularly polarized light. Theodosiou [9] calculated minima in the emission oscillator strengths of the Rydberg states of the alkali-metal atoms that appeared only in the *D* states of potassium and *F* states of cesium. Sandner *et al*. [10] measured the photoionization in the vicinity of the minimum cross section from the ground state of the atomic potassium. Davenport *et al*. [11] calculated the photoionization cross section and oscillator strengths for the strongest bound-bound transitions of Li_2, Na_2, and K_2 using the multiple scattering theory. They reported that the Cooper minimum in Na_2 was broadened considerably over the atomic Na because the matrix elements for the σ and π channels pass through zero at different photon energies. Duncan *et al*. [12] measured the photoionization cross section of potassium, K, using a high-intensity $\cong$1 kW/cm^2 nanosecond laser at 266 nm wavelength and reported that the single-photon excitation gets suppressed due to the Cooper minimum in the continuum.

Petrov *et al*. [13] reported the effect of the polarization of the atomic core by the outer electron on the near-threshold photoionization of the excited alkali-metal atom. Beterov *et al*. [14] reported the anomalous behavior for the *S* series, the rate of high-lying Rydberg levels as a function of *n* using blackbody radiations which was found to be similar to the occurrence of the Cooper minimum in the discrete spectrum. Sahoo and Ho [15] studied the plasma screening effect to

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uncover the Cooper minimum in the photoionization cross sections from the ground state of the Li atom embedded in the Debye plasma environment. Recently, Zatsarinny and Tayal [16] investigated the photoionization of atomic potassium from the $4s$ ground state and ns ($n = 5-7$) excited states of potassium atoms using a Dirac-based B-spline R-matrix method. They obtained excellent agreement with experimental data for the cross sections of the $4s$ photoionization, including an accurate description of the near-threshold Cooper-Seaton minimum. More recently Betrov *et al.* [17] described the structure of the Cooper minima in the transition probabilities and photoionization cross sections for the low excited and Rydberg nS, nP, nD, and nF states of alkali-metal atoms using the Coulomb approximation and a quasiclassical model.

To our knowledge, there are hardly any experimental data on the photoionization cross section from the $7s\ ^2S_{1/2}$ excited states of potassium despite the availability of extensive theoretical calculations for the ns ($n = 4-7$) excited sates of potassium [16]. In our present work we have validated experimentally the theoretically predicted Cooper minimum in the photoionization cross sections of the $7s\ ^2S_{1/2}$ excited state of potassium. This work is in continuation of our experimental studies [18–29] on the measurements of the photoionization cross sections for the excited states of alkali-metal and alkaline atoms. We report here the measurements of the photoionization cross sections from the $7s\ ^2S_{1/2}$ excited state of potassium at four different ionizing lasers wavelengths, about 3 eV above the first ionization threshold using the two-step photoionization saturation techniques in conjunction with a time-of-flight (TOF) mass spectrometer. The measured values are found to be in excellent agreement with the theoretical calculations.

II. EXPERIMENTAL DETAILS

The experimental setup to measure the photoionization cross sections from the $7s\ ^2S_{1/2}$ state of potassium is very similar as described in our earlier studies on the measurements of the photoionization cross section of potassium [18], lithium [20–22], magnesium [23], and barium [24]. We have employed the two-photon–two-step excitation and ionization technique using two dye lasers pumped by a common Nd:YAG laser. About 70% of the available power of the second-harmonic generation (SHG) (532 nm) of the Nd:YAG laser was used to pump the TDL-90 (Quantel) dye laser. The LDS-698 and DCM-640 dyes mixed in a ratio of 2:3 dissolved in methanol were used to charge the dye laser that was tuned at 660.59 nm. The bandwidth of the dye laser was $\leqslant 0.1$ cm^{-1} and the output laser energy was about 300 μJ. The dye laser was tuned at 660.59 nm to promote the potassium atoms from the $4s\ ^2S_{1/2}$ ground state to the $7s\ ^2S_{1/2}$ excited state via the two-photon excitation process. For the second step, we have used four ionizing lasers of wavelengths 1064, 700, 532, and 355 nm to photoionize the potassium atoms from the $7s\ ^2S_{1/2}$ state. The ionizing laser wavelength of 700 nm was prepared using a Hana-type dye laser charged with LDS-698 dissolved in methanol and pumped by about 30% remaining power of the SHG of the Nd:YAG laser. Two of the ionizing laser wavelengths, 1064 and 532 nm, are the fundamental and the second harmonic of the same Nd:YAG laser. Since we have no option to use third-harmonic generation (THG) of the Nd:YAG laser therefore, the 355-nm ionizing laser wavelength was achieved from the THG of another Nd:YAG laser by synchronizing with the first Nd:YAG laser using a trigger circuit and a digital delay pulse generator (SRS DG 535).

A spectroscopically pure potassium sample was loaded in the oven placed in a vacuum chamber, and then evacuated up to $\sim 10^{-6}$ mbar. The oven was heated at a constant temperature $\approx$465 K. The temperature of the oven was maintained within ± 1 K during the experimentation. A well-collimated atomic beam of potassium was produced that traveled from the lower to the upper part of the vacuum chamber and passed through the center of the ionization (extraction) region of the TOF mass spectrometer where it interacts orthogonally with the exciting and the ionizing laser beams. The exciting laser beam having a diameter of 4.5 mm filled the entire atomic beam diameter in the ionization-extraction region of the TOF mass spectrometer. The ionizer laser beam was focused at the center of the above-mentioned region with a planoconvex lens of focal length 50 cm. It enabled us to make the ionizing volume as nearly cylindrical as possible over the ion collection length, which was equal to the calculated diameter (4 mm) of the atomic beam in the interaction region. Both the laser pulses were linearly polarized with the parallel polarization axis. Since both the laser beams originate from the same Nd:YAG laser, we nevertheless confirmed the temporal overlap with a photodiode. Interaction of the laser beams with the atomic beam produces potassium ions that were detected by a Channeltron. The TOF detector produces voltage signals of potassium ions across a 50-Ω resistor, which was converted to the number of ions produced. The detector's linearity is very important while measuring the photoionization cross section. An optimum operating voltage was determined for the Channeltron corresponding to the maximum available laser intensity in order to get the true photoion signals. The temporal overlapping of the exciting and the ionizing lasers is also very essential; it was adjusted using an optical delay line and confirmed with a PIN photodiode. The intensity of the ionizing laser was varied by inserting neutral density filters (Edmund Optics), and on each insertion the energy was measured by an energy meter (R-742).

III. RESULTS AND DISCUSSION

The photoionization cross sections have been measured in the region of the Cooper minimum from the $7s\ ^2S_{1/2}$ excited state above the first ionization threshold of potassium. The cross sections have been measured by employing the two-step, two-photon excitation and ionization technique using a TOF mass spectrometer. The excited $7s\ ^2S_{1/2}$ state was populated via two-photon excitation from the $4s\ ^2S_{1/2}$ ground by adjusting the dye laser at 660.59 nm. The laser light is linearly polarized having the polarization vector in the z direction. Since both the $4s\ ^2S_{1/2}$ and $7s\ ^2S_{1/2}$ levels possess $m_J = 1/2$ states, the linearly polarized laser light transitions between these two states follow the $\Delta m_J = 0$ selection rule. In the second step, the atoms from the $7sS_{1/2}$ level are ionized at four laser wavelengths, 1064, 700, 532, and 355 nm. The ionizing lasers are also linearly polarized and their polarization direction

is kept parallel to that of the first-step dye laser. According to the dipole transition selection rules in LS coupling for two-photon excitations ($\Delta\ell = 0 \pm 2, \Delta S = 0, \Delta J = 0, \pm 2$ and $\Delta L = 0, \pm 2$) the first-step excited levels are

$$4s\,^2S_{1/2} \xrightarrow{2\hbar\omega} ns\,^2S_{1/2}.$$

The ionization from the $7sS_{1/2}$ level approaches the εp continuum channels via single-photon excitation:

$$4s\,^2S_{1/2} \xrightarrow{2\hbar\omega} 7s\,^2S_{1/2} \xrightarrow{\hbar\omega} \varepsilon p\,^2P_{1/2,3/2}.$$

Thus, we can promote atoms from the ground state to the continuum channels using the two-photon, two-step excitation (photoionization) technique. As the transition in the first step is saturated, therefore the time-dependent number densities in the ground and the excited states are equally distributed. In the second step, the atoms from the $7s$ excited state are photoionized by the ionizing laser. Thus the ionization rate from the excited state which is related to the photoionization cross section, the intensity, and the energy of the ionizing laser is represented by the relation

$$R_{\text{ion}} = \frac{I(t)\sigma}{\hbar\omega}. \tag{1}$$

Here σ (cm^2) is the cross section of photoionization from the excited state to the continuum channels at the ionizing laser wavelength, I (W/cm^2) is the time-dependent intensity, and $\hbar\omega$ (J) is the photon energy of the ionizing laser. The rate equations involving the number densities in the ground state, excited state, and the ionization channels have been solved to derive a relation for the number of ions produced as a function of the intensity of the ionizing laser. This method was originally suggested by Ambartzumian *et al.* [30] and Heinzmann *et al.* [31] and has been used and described by a number of researchers [20–37]; it has been extensively used for the measurement of photoionization cross sections of lithium isotopes [20–22] and of the excited states of alkali-metal and alkaline earth metals [20–34]. Saturation is explained by the fact that as the intensity of the ionizing laser is increased, the ion signal keeps on increasing until the point at which any further increase in the ionizing laser intensity will not increase the ion signal. At this point the total ionization of the excited atoms takes place and saturation in the ion signal is achieved. The rate equations solution yields a relation between the voltage signal and the photoionization cross section [24–27] as

$$\text{Voltage signal} = \frac{eN_0VR}{\Delta t}\left[1 - \exp\left(-\frac{\sigma E}{2\hbar\omega A}\right)\right]. \tag{2}$$

Here, Δt (s) is the full width at half maximum of the photoion's signal, e (C) is the electronic charge, R (Ω) is the load resistor, V (cm^3) is the laser-interaction volume, N_0 (cm^{-3}) is the number density of atoms in the excited state, A (cm^2) is the area of cross section of the ionizing laser, $\hbar\omega$ (Joule) is the energy of the ionizing photon, E (J) is the energy of the ionizing laser pulse, and σ (Mb) is the photoionization cross section of the excited state. All the quantities in this relation are known except N_0 and σ which can be extracted by the least-squares fit of Eq. (2) to the experimental data. The above equation shows that the ion signal gets saturated if the energy density of the ionizing laser is infinite.

Accurate determination of the photoionization cross section and number density demands accurate measurements of the ionizing laser energy density as well as the characterization of the spatial profiles of both the exciting and the ionizing laser pulses in the interaction region. An aperture of diameter ≈3.8 mm was placed in the path of the exciting laser whereas another aperture of smaller diameter ≈2.6 mm was placed in the path of the ionizing laser. Since the diameter of the ionizing laser is kept smaller than that of the exciting laser beam it therefore reduces the problems associated with the spatial overlap of the laser beams in the interaction region. A lens of long focal length 50 cm was also used in the path of the ionizing laser to meet the power requirements for saturation. The measurement of the spatial profiles of both the exciting and the ionizing laser beams was carried out with two beam splitters placed before the entrance to the interaction region by scanning a PIN photodiode across their diameters. The intensity distribution of both the laser beams was found to be Gaussian. Furthermore, by detecting ions rather than photoelectrons, we avoided the normalization problems arising from the angular distribution of the electrons [10].

In Fig. 1, we present the photoionization data for the photoionization from the $7s\,^2S_{1/2}$ state of potassium using the ionizing laser wavelength at 1064 nm. It is evident from the figure that the change in the signal height with the ionizing laser energy (intensity) almost remains constant and does not approach the saturation state at the given laser energy (intensity). The solid squares are the experimental data points; the error bars show ≈5% uncertainty. The solid line is the least-squares fit of Eq. (2) to the experimental data that yields the value of the cross section σ as $(9.78 \pm 0.14) \times 10^{-2}$ Mb at the 1064 nm ionizing laser wave lengths and the value of the number density N_0 as 2×10^{11} cm^{-3}. We repeated this experiment thrice and recorded the ionization signals from

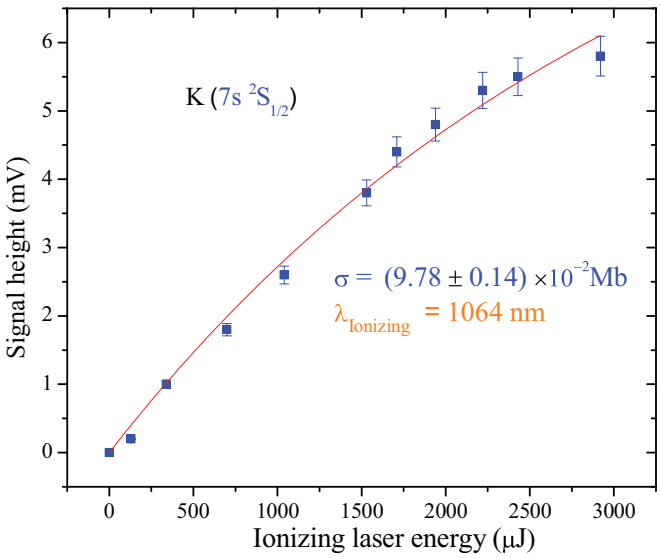

FIG. 1. (Color online) Photoionization cross-section data from the $7s\,^2S_{1/2}$ state of potassium as a function of ionizing laser energies using 1064-nm ionizing laser wavelength. The solid line that passes through the experimental points is the fit of Eq. (2).

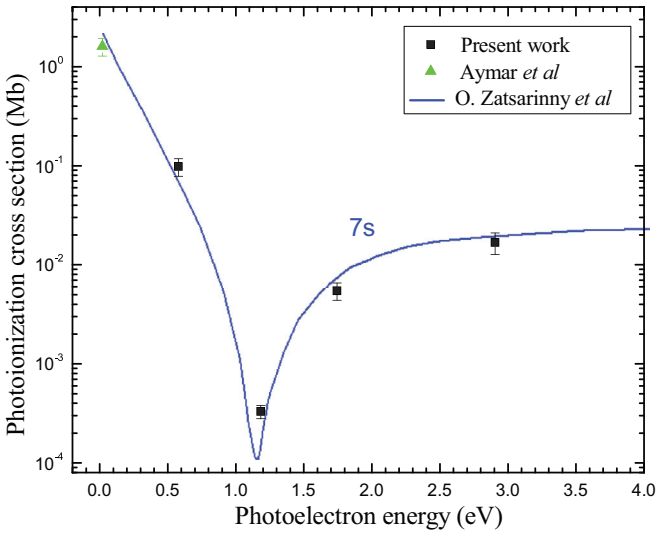

FIG. 2. (Color online) Comparison of the experimentally determined absolute values (points) of the photoionization cross sections (Mb) and the theoretical calculations (line) (Zatsarinny and Tayal [16]) for the $7s\ ^2S_{1/2}$ state as a function of the excess photoelectron energy.

the $7s\ ^2S_{1/2}$ state using the ionizing laser at 700, 532, and 355 nm. It is notable that a complete saturation of ionization as Eq. (2) illustrates is possible only if the ionizing laser energy (intensity) approaches infinity. The fitting procedure of the curves of Eq. (2) to the experimental data points yields the photoionization cross sections from the $7s\ ^2S_{1/2}$ excited state as $(3.30 \pm 0.49) \times 10^{-4}$,$(5.47 \pm 0.82) \times 10^{-3}$, and $(2.06 \pm 0.30) \times 10^{-2}$ Mb at the ionizing laser wavelengths 700, 532, and 355 nm, respectively. The main sources of errors in the measurement of the cross section are the uncertainties in the laser energy measurements and the cross-sectional area of the ionizer laser beam in the interaction region. The uncertainties in the energy measurements are due to the energy meter ($\cong \pm 3\%$) and pulse-to-pulse variations in the laser energy ($\cong \pm 5\%$). Additional uncertainty comes from the area measurement of the spatial beam profile of the ionizer laser beam ($\cong \pm 10\%$). The maximum overall uncertainty in the determination of the absolute cross section is $\cong 15\%$.

In Fig. 2 we present our experimental data of the measured photoionization cross sections from the $7s\ ^2S_{1/2}$ state at four different ionizing wavelengths, 1064, 700, 532, and 355 nm. The solid line following the data points shows the theoretical calculations by Zatsarinny and Tayal [16]. Our measurements clearly show a minimum in the photoionization cross section at $\cong 1.25$ eV above the first ionization threshold. The minimum cross section $\cong 10^{-4}$ Mb has been obtained using the 700-nm ionizing laser wavelength. A comparison of the photoionization cross sections measured from the $7s\ ^2S_{1/2}$ excited states with the existing experimental and theoretical values is tabulated in Table I.

As shown by Fig. 2, our measured values are in good agreement with the theoretical values reported by Zatsarinny and Tayal [16]. Considering 15% uncertainty in the present measurements it is evident that the minimum values of the cross section are in excellent agreement with the theoretical calculations. Zatsarinny and Tayal [16] inferred a huge difference of several orders between the experimentally determined photoionization cross section from the $7s$ state by Amin *et al.* [19] and their calculations at this energy. We take this opportunity to correct the earlier measurements of the photoionization cross section from the $7s$ state by Amin *et al.* [19]. Amin *et al.* [19] used a single dye laser and a thermionic diode as an ion detector. The term energy of the $7s$ level is 30 274 cm^{-1} which lies about 4000 cm^{-1} below the first ionization threshold. This level was populated via two-photon excitation from the ground state; single-photon energy is 15 137 cm^{-1}. By adding the energy of the third photon the total energy becomes 5.63 eV which is about 1.29 eV above the ionization threshold. Thus Amin *et al.* [19] came to a conclusion that they had measured the photoionization cross section at 1.29 eV above the ionization threshold. In fact, the experimental results were the resonant two-photon ionization from the $7s$ state. In their experiment the collisional ionization was dominant which made them to come to a conclusion that they have measured the cross section above the threshold, which was in fact the collisional excitation from the $7s$ level. In the present work, we have rectified the problem of collisional ionization and employed an atomic beam apparatus coupled with a time-of-flight mass spectrometer so that only the photoions are registered. We also ensured the two-step excitation (ionization) process using two dye lasers delayed by about a few nanoseconds. In the absence of the ionizing laser, there was no ionization signal and if the first-step laser was off resonance then again the ionization signal disappeared.

In our earlier work, we have measured the photoionization cross section from the $4p$ level at 355 nm [19] and from the $6p$ level at four different ionizing laser wavelengths [18]. The photoionization cross section from the $4p$ level has a

TABLE I. Comparison of the photoionization cross sections measured from the $7s\ ^2S_{1/2}$ excited state of potassium with the theoretically calculated values.

Present work (experimental)			Previous work (calculated)[a]	
Cross section σ (Mb)	Ionizing laser (nm)	Excess energy (eV)	Cross section σ (Mb)	Ionizing laser (nm)
$(9.78 \pm 0.14) \times 10^{-2}$	1064	0.578	6.86×10^{-2}	1064
$(3.30 \pm 0.49) \times 10^{-4}$	700	1.184	2.23×10^{-4}	701
$(5.47 \pm 0.82) \times 10^{-3}$	532	1.743	8.50×10^{-3}	532
$(2.06 \pm 0.30) \times 10^{-2}$	355	2.905	2.14×10^{-2}	355

[a]The theoretical results are the approximate values obtained from the published curves of data reported by Zatsarinny and Tayal [16].

lower value at the ionization threshold, achieves a higher value at about 0.5 eV above the threshold, and then decreases monotonically [13]. The photoionization cross section from the $6p$ level shows an increasing trend from the ionization threshold whereas a Cooper minimum in the photoionization from the $4s$ level has been studied by Sandner *et al*. [10]. The theoretical calculation for the photoionization cross section from the $5p$ level also predicts an increasing trend up to 1 eV above the threshold and then a monotonic decrease. It will be interesting to study the cross section from this level as well to validate the theoretical predictions. We are planning to explore this part of the spectrum in the near future.

In conclusion, the photoionization cross section from the $7s\ ^2S_{1/2}$ excited state of potassium measured up to 2.9 eV above the first ionization threshold is found to be higher near the ionization limit and then decreases rapidly to its minimum value up to the Cooper minimum at about 1.25 eV. With further decrease in the ionizing laser wavelengths the cross section increases, first sharply, and then slowly attains its asymptotic value. The absolute values of the cross sections determined in the present work lie almost on the theoretically calculated curve. Thus we have been able to verify the existence of the Cooper minimum in the photoionization from the $7s$ state of potassium as theoretically predicted by Zatsarinny and Tayal [16].

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