

Mechanisms of Plasma Formation in Potassium Vapor Excited by Nanosecond Resonant Laser PulsesKholoud A Hamam^{1*}, and M. A. Mahmoud²¹Department of Physics, Faculty of Science, King Abdulaziz University, Jeddah, KSA.²Physics Department, Faculty of Science, Sohag University, 82524-Sohag, Egypt.*Corresponding author: Email: khamam@kau.edu.sa, kholoud_797@hotmail.com

Abstract: We have studied theoretically formation of molecular ion K_2^+ and the atomic ion K^+ which are created in laser-excited potassium vapor at the first resonance transition, 4S - 4P. A set of rate equations, which describe the temporal variation of the electron energy distribution function (EEDF), the electron density, the population density of the excited states as well as the atomic K^+ and molecular ion K_2^+ , are solved numerically. The calculations are carried out at different laser energy and different potassium atomic vapor densities under the experimental conditions given by Amin et al. [19]. In this experiment, the potassium atomic density was varied in the range from 3.5×10^{15} to $9 \times 10^{15} \text{ cm}^{-3}$; the vapor temperature was 540 K° , and the laser repetition rate was 10 Hz. The intensity of the exciting laser beam is 0.5 mJ, while the intensity of the ionizing laser beam was varied in the range from 0.1 to 1 mJ, and the laser beam diameter was varied in the range 230 - 290 mm. The numerical calculations of the electron energy distribution function (EEDF) show that a deviation from the Maxwellian distribution due to the super-elastic collisions effect. In addition to the competition between associative ionization (4p-4p), and Molnar -Hornbeck ionization processes for producing K_2^+ , the calculations have also shown that the atomic ions K^+ are formed through the Penning ionization and photoionization processes. These results are found to be consistent with the experimental observations.

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1. Introduction

In recent years, the study of collisional ionization, and excitation energy transfer processes in a laser-excited alkali vapor have been the subject of numerous studies [1-8]. This phenomenon attracted the attention of many researchers due to its importance in different fields in the applied areas of science and technology such as gas lasers, astrophysics, photochemistry and controlled thermonuclear [4-8]. By studying the physics of chemical-ionization processes, researchers are exploring the fundamental assumptions of the theory of atomic collisions and the physics of low-temperature plasmas and hence the possibility of new phenomena and new properties [7-9].

On the other hand, laser-induced or laser-assisted collisional ionization technique has been extensively studied ever since high-intensity pulsed lasers have been developed. This technique has played a vital role in coupling laser energy into vapor in applied science. Laser-plasma mechanism is important in various studies like laser fusion, energy conversion, optical switching, X-ray laser development, laser heating magnetically confined plasmas, and others. In pure science, however, laser breakdown is involved in studies like ion spectroscopy [10]. Then again resonant

heating of free atoms may also be important in resonant laser ablation and resonantly laser-induced breakdown spectroscopy [11-13].

A theoretical model was proposed by Measures [14-17] to explain laser ionization based on resonance saturation (LIBORS). The model assumes that the laser saturates the resonance level and initially provides seed electrons by various energetic processes. Besides, the reduction of the ionization energy of the laser excited atoms leads to the electron growth. These seed electrons are then heated up within the medium as a result of super-elastic collisions which act to quench the overpopulated resonance level. Those heated electrons continue to collisionally excite electrons to high-lying states that can be further ionized in the laser field or by direct collisional ionization producing secondary electrons. Runaway collisional ionization of the upper levels occurs until super-elastic heating can no longer balance collisional cooling, and the electron temperature falls. It was shown that there exists a competition of several ionization reactions: two-photon ionization of resonant states, associative ionization of resonant states, direct ionization by electron impact of ground and the resonant state, stepwise ionization by electron impact, and

photoionization from high -lying excited states by resonance radiation.

This work aims to modify the model given in [18, 19] which previously under took the phenomenon associated with laser-induced ionization of metallic vapors based on resonance saturation. The modified model is applied to investigate the experimental measurements which are carried out by Amin et al., [20] to study nanosecond pulsed Nd: YAG laserpumping potassium vapor leading to their two-step excitation and ionization and plasma generation. The computations are performed to analyze; a) excitations and ionizationkineticsb) electron and ions (K^+ and K_2^+) densitydependence on laser power and potassium vapor density. To validate the applied model comparison is performed between the calculated and experimentally measured atomic ion density corresponding to the considered experimental condition.

2. Theoretical model

The main feature ofthe developed LIBORSmodel is that the energy, necessary to ionize a working vapor

$$\begin{aligned} \frac{dN(4s)}{dt} = & N(4p)(R_{21} + A_{21}) - N(4s)R_{12} + N(4p) \int n_e(\epsilon)k_{21}(\epsilon) d\epsilon - N(4s) \int n_e(\epsilon)k_{12}(\epsilon)d\epsilon \\ & + N(4p)N(n)k_{PI} + \frac{1}{2}N^2(4p)k_{EP} - N(n)N(4s)k_{HMI} - N(4s) \int n_e(\epsilon)k_{1C}(\epsilon)d\epsilon \quad (1) \end{aligned}$$

The rate of change of the population density of the resonance level (4P) is given by:

$$\begin{aligned} \frac{dN(4p)}{dt} = & N(4s)R_{12} - N(4p)(R_{21} + A_{21}) - N(4p) \int n_e(\epsilon)k_{21}(\epsilon)d\epsilon + N(4s) \int n_e(\epsilon)k_{12}(\epsilon)d\epsilon - \frac{1}{2}N^2(4p)k_{AI} \\ & - N(4p)N(n)k_{PI} - \frac{1}{2}N^2(4p)\sigma_{PI}vF - \frac{1}{2}N^2(4p)k_{EP} - N(4p)\sigma_{2C}^{(2)}F^2 \\ & - N(4s) \int n_e(\epsilon)k_{2C}(\epsilon)d\epsilon \quad (2) \end{aligned}$$

The rate of change of the population density of the highly excited states ($n > 4P$) is shown as:

$$\begin{aligned} \frac{dN(n)}{dt} = & \sum_{n>m} n_e(\epsilon) N(n)k_{nm}(\epsilon) - \sum_{m>n} n_e N(n)k_{mn}(\epsilon) - \sum_{n>m} A_{nm}N(n) - \sum_n n_e(\epsilon) N(n)k_{nc}(\epsilon) \\ & - \sum_n N(4p) N(n)k_{PI} + \frac{1}{2}N^2(4p)k_{EP} - \sum_n N(n) N(4s)k_{HMI} - \sum_{n>2} N(n)\sigma_{nc}^{(1)} F \\ & + N_k + n_e(\epsilon) \sum_n [n_e(\epsilon)k_{cn}(\epsilon) + k_{RD}(\epsilon)] \quad (3) \end{aligned}$$

The growth rate of the atomic and molecular ion is given by:

$$\begin{aligned} \frac{dN(K^+)}{dt} = & \sum_n N(4p) N(n)k_{PI} + \sum_n n_e(\epsilon) N(n)k_{nc}(\epsilon) + N(4p)\sigma_{2C}^{(2)}F^2 + \sum_{n>2} N(n)\sigma_{nc}^{(1)} F + \frac{1}{2}N^2(4p)\sigma_{PI}vF \\ & - N_k + n_e(\epsilon) \sum_n [n_e(\epsilon)k_{cn}(\epsilon) + k_{RD}(\epsilon)] \quad (4) \end{aligned}$$

$$\frac{dN(K_2^+)}{dt} = \frac{1}{2}N^2(4p)k_{AI} + \sum_n N(n) N(4s)k_{HMI} \quad (5)$$

atom E_{1c} should be greater than twice the energy between the ground state, and the first excited (resonance) state, E_{12} , andless than three times this energy, i.e., $2E_{12} < E_{c1} < 3E_{12}$. The alkali metals exhibit this property. The present work is devoted to investigating plasma generation of potassium vapor induced by pulsed laser radiation with photon energy $h\nu = E_{12}$.

2.1 The physical processes

Following to the energy level diagram of potassium atom shown in Figure (1), in the present analysis, the illumination of potassium vapor with a tuned pulsed laser source leads to some collisional and radiative processes similar to those given in our previous papers [21, 22].

2.2 Method of calculations

The rate of change of the population density of the atomic states shown in Figure (1) beside the formed atomic and molecular ions are given as follows.

The rate of change of the population density of the ground state (4S) is presented as:

The equation which represents the electron energy distribution function is given by:

$$\begin{aligned} \frac{dn_e(\varepsilon)}{dt} = & \sum_{m>n} n_e(\varepsilon) N(m) k_{nm}(\varepsilon) - \sum_{m<n} n_e(\varepsilon) N(n) k_{mn}(\varepsilon) + \sum_n n_e(\varepsilon) N(n) k_{nc}(\varepsilon) + \sum_n N(4p) N(n) k_{pI} \\ & + N(4p) \sigma_{2c}^{(2)} F^2 + \sum_{n>2} N(n) \sigma_{nc}^{(1)} F + \frac{1}{2} N^2(4p) \sigma_{pI} v F + \frac{1}{2} N^2(4p) k_{AI} + \sum_n N(n) N(4s) k_{HMI} \\ & - N_{K^+} n_e(\varepsilon) \sum_n [n_e(\varepsilon) k_{cn}(\varepsilon) + k_{RD}(\varepsilon)] \quad (6) \end{aligned}$$

The normalization conditions are:

$$N_0 = \sum_n N(n) + N(K^+) \quad (7)$$

$$\int_0^\infty n_e(\varepsilon) \varepsilon^{\frac{1}{2}} d\varepsilon = 1, \int_0^\infty n_e(\varepsilon) d\varepsilon = N_e \quad (8)$$

Where $N(4s)$, $N(4P)$ and $N(nl)$ are the population density of levels $4s, 4P$ and nl respectively where nl is any excited level $> 4p$ (cm^{-3}). K^+ represents the density of atomic ions (cm^{-3}). K_2^+ represents the density of molecular ions (cm^{-3}). N_0 is the density of K atomic vapor (cm^{-3}). $n_e(\varepsilon)$ refers to the density of free electrons with an energy ε and $\varepsilon + \Delta\varepsilon$ (cm^{-3}). R_{12} refers to the stimulated absorption rate for the transition $4s - 4p$ level (sec^{-1}). R_{21} signifies the stimulated emission rate for the transition $4p - 4s$ (sec^{-1}). A_{nm} represents the Einstein coefficient for spontaneous emission for the transition $n \rightarrow m$ (sec^{-1}), where $n > m$. k_{21} represents the de-excitation rate coefficient for the $4p \rightarrow 4s$ atomic transition ($\text{cm}^3 \cdot \text{sec}^{-1}$). k_{12} represents the excitation rate coefficient by electron energy $\varepsilon \geq \varepsilon_x$ for the atomic transition $4s \rightarrow 4p$ ($\text{cm}^3 \cdot \text{sec}^{-1}$). $\sigma_{nc}^{(1)}$ represents the single photon ionization cross-section (cm^2). $\sigma_{2c}^{(2)}$ represents the two-photon ionization cross-section (cm^{2K}). F photon flux (photon $\text{cm}^{-2} \text{sec}^{-1}$). v represents the relative velocity of the free electrons (cm sec^{-1}).

k_{1c} represents electron impact ionization rate coefficient for level $4s$ ($\text{cm}^3 \cdot \text{sec}^{-1}$). k_{2c} represents electron impact ionization rate coefficient for level $4p$ ($\text{cm}^3 \cdot \text{sec}^{-1}$). $k_{nc}(\varepsilon)$ characterizes rate coefficient of electron impact ionization of any higher level n ($\text{cm}^3 \cdot \text{sec}^{-1}$). k_{cn} denotes the three-body recombination rate coefficient to any level n ($\text{cm}^6 \cdot \text{sec}^{-1}$). k_{nm} represents the de-excitation rate coefficient ($\text{cm}^3 \cdot \text{sec}^{-1}$) of the transition $n \rightarrow m$, through excited atoms collision with low energy electrons. $k_{mn}(\varepsilon)$ refers the excitation rate coefficient ($\text{cm}^3 \cdot \text{sec}^{-1}$) for the transition $m \rightarrow n$ by high energy electrons. k_{RD} denotes the radiative recombination rate coefficient ($\text{cm}^3 \cdot \text{sec}^{-1}$). k_{pI} represents Penning ionization rate coefficient for level n ($\text{cm}^3 \cdot \text{sec}^{-1}$). k_{EP} characterizes the rate coefficient of the energy pooling collisions ($\text{cm}^3 \cdot \text{sec}^{-1}$). k_{AI} represents the association ionization rate coefficient ($\text{cm}^3 \cdot \text{sec}^{-1}$). k_{HMI} represents the Horn-beck Molnar ionization rate coefficient ($\text{cm}^3 \cdot \text{sec}^{-1}$). σ_{pI}

represents Penning ionization cross section (cm^2). σ_{pI} represents induced Penning ionization cross section (cm^2).

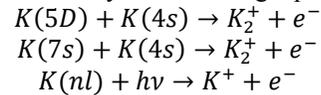
Note that the factor $\frac{1}{2}$ appeared in the terms express the energy pooling, associative ionization, and induced Penning ionization mechanisms are added to correct for possible double counting of each colliding pair of identical particles [23].

3. Results and Discussion

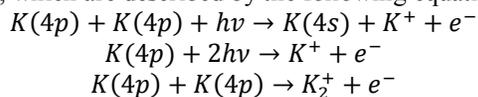
In this section we will present and discuss the results of calculation of the first resonance ionization ($4S-4P$) of potassium vapor under the experimental conditions given in [20]; where a pulsed dye-laser beam is used. The dye laser having line width $\leq 0.3 \text{ cm}^{-1}$ with repetition rate equals to 10 Hz, pulse duration $\approx 6.0 \text{ ns}$ and energy 135mJ per pulse. The intensity of the exciting laser beam was fixed $\approx 0.5 \text{ mJ}$, whereas the intensity of the ionizing laser beam was varied from a minimum value 0.1mJ to 1mJ. The laser energy varied from 0.1 mJ to 1mJ, using potassium vapor density varying in a range from 3.57×10^{15} to $8.94 \times 10^{15} \text{ cm}^{-3}$.

3.1 Variation of electron energy distribution function with time

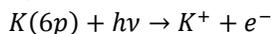
The time evolution of the electron energy distribution function at different time intervals shown in figure (2). These calculations enabled us to study the time evolution of the processes responsible for electrons seeding and growing. This can be seen from the noticeable increase of the height of the peaks (A, B, C, D, E, and F) corresponding to energies 0.08, 0.5, 0.73, 1.09, 1.48, 1.72eV respectively. The height of the peaks increases noticeably as the time increase. A correlation between the position of the peak and electron energy specify exactly the physical process responsible for each peak. The energy of the electrons produced in peak A which is centered at mean energy $\varepsilon = 0.08 \text{ eV}$ created by Horn-beck Molnar ionization of $7s$ and $5d$ and single photoionization of $3d$ and $5s$, which are described by the following equations:



While the peak B which is attributed to electrons produced by laser induced-Penning ionization, two photo ionization and Associative ionization of the 4p state, which are described by the following equations:

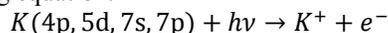


The peak C corresponds to single photo ionization of the 6p state, which is described by the following equation:

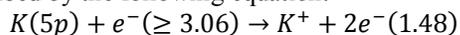


On the other hand, the fast transition from 5 to 20 ns; might be attributed to the high rate of the energy pooling collision processes which populate the highly excited states like 5p, 4d, 6s. The main population source of these excited states is the population of the 4P state which results by laser pumping leading to the 4S - 4P transition. Energy pooling mechanism then plays a significant role in populating the highly excited states.

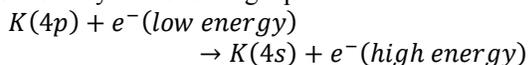
The peak D corresponds to single photoionization of 4p, 5d, 7s, and 7p which are described by the following equation:



While the peak E is attributed to electron impact ionization of highly excited states (5p), which are described by the following equation:



The remaining peaks are attributed to the superelastic collisions. This could be noticed from the clear replication of the characteristics of the peaks appeared after the peak E over the high energy region. The first group of these peaks attributed to the first superelastic collision between electrons produced by associative ionization, Hornbeck-Molnar ionization, photoionization, electron impact ionization or Penning ionization and then heated up through the collision transfer of energy of K (4p) atoms, which are described by the following equation:



Also, the second group of these peaks shown in figure (4-2) attributed to the second super-elastic collision. The disappearance of the peak F at t=1 and 5ns may be attributed to the fact that the electrons having energies above (1.7 eV) can easily collide with the K (4p) excited states resulting in its ionization.

At this point, we can conclude that the ionization of K atom based on laser saturation is mainly produced through single, two photoionizations, induced Penning ionization, associative ionization, and Hornbeck-Molnar ionization. The energy pooling is responsible for the population of the highly excited states, and the successive superelastic collisions are responsible for the heating of electrons resulting on rapid ionization of the medium [16,18,24, 25].

3.2 Time evolution of atomic ions K⁺

The growth rate of the atomic ions K⁺ as a function of time for different values of potassium vapor density is indicated in the figure (3). As shown in the figure, at the early stages of the interaction, the density of atomic ions shows a fast increase up to 10ns. This behavior can be explained by the fact that the main process for producing atomic ions, K⁺, is the photoionization, Penning ionization, laser-induced Penning ionization and electron-impact ionization of the ground and excited atomic states of K atoms. The energy pooling processes of 4p state populate the other highly excited states which can easily ionize through photo or Penning ionization processes. These processes are more effective at the early stages of the plasma formation since its occurrence depends on the growth rate of the population density of K (4p) and any of the highly excited states. Then electron can gain enough energy through one or more of successive super elastic collisions to ionize excited atoms through electron impact ionization processes. From the curves, we note that the behavior of atomic ions variation does not change with increasing the potassium vapor density. The slow growth rate observed in figure (3) after time 10ns is due to some processes which lead to a decrease of the atomic ions population density like the radiative recombination and the three body recombination processes unlike the atomic ions creation processes [18, 20,26].

3.3 Time evolution of molecular ions K₂⁺

The growth rate of the molecular ions K₂⁺ as a function of time is illustrated in figure (4) for different values of potassium vapor density. From this figure we can see that the K₂⁺ density increases rapidly during the period, 0.1 nsec up to 10 nsec followed by a slow increase up to 10 nsec for the different values of the potassium vapor density. The explanation of the linear growth can be described as follows: during the early stages, the molecular ions are created at a high rate through Associative and Horn-beck Molnar ionization. These 4p atoms created as soon as laser illumination, the density of the 4p atoms is high, so the creation of the molecular ions will be fast in the early stages of the plasma formation. After this time the K₂⁺ density shows a slow increase during the late stages of the irradiation time. This slowing in molecular ions production is due to the decrease in the density of 4p, 5d and 7s atoms, where most of these atoms create molecular ions through Associative and Horn-beck Molnar ionization [5,27].

The dependence of molecular ions density on potassium vapor density at different irradiation times and laser energy equals to 0.1 mJ is shown in figure (5). From this figure, we note a fast increase of the molecular ions density with the increase of potassium vapor density. This behavior can be interpreted as the

result of the increase of the 4p atoms density with the increase of the potassium vapor density. Many of these 4p atoms converted to molecular ions through Associative ionization [7,18, 28, 29]. Also, these 4p states may be excited to the 5d and 7s states (through energy pooling collisions) which can create molecular ions through Hornbeck-Molnar ionization. This figure confirms the previous results which are obtained in the previous figure (4).

3.4. Comparison with the experimental results

Comparison between the calculated atomic ions density under the experimental conditions given in [20] and the measured ones is shown in figure (6). This figure illustrates the calculated atomic ions density for a time interval 6 ns versus the ionizing laser energy over a range 0.1 mJ to 1.0 mJ, the laser beam is adjusted at wavelengths 770.1nm for the K (4p $^2P_{1/2}$) state and 766.7nm for the K (4p $^2P_{3/2}$) state. From this figure, it can be seen that good agreement between the calculated and the measurement values. The coincidence of both values confirms the experimental observation which indicated that as the

intensity of the ionizing laser increases, the ion signal increases up to a certain value very swiftly and then changes slowly.

We obtained results are interpreted on the bases that the interaction involves successive absorption of the laser photons by the K (4s) state leading eventually to the population of the resonantly photo-excited K (4p $^2P_{1/2}$) and K (4p $^2P_{3/2}$) states. As a result of this population, these K (4p) atoms undergo many interactions such as; associative ionization, Penning ionization, Hornbeck-Molnar, and photoionization. Moreover, These atoms can decay to the ground state K (4s) after 26.1 ns (the lifetime of the 4p $^2P_{1/2}$ state) or 25.8 ns (the life time of the 4p $^2P_{3/2}$ state). Collision of two K (4p) atoms through energy pooling interaction leading finally to the population of the highly excited states. Super elastic collisions and photoionization processes of these formed highly excited states are the processes responsible for atomic ionization and in turn positive atomic ions formation [5,7,13].

Table (1): Some cross sections of potassium atom.

Process	Equation	Cross Section	Reference
Laser induced-penning ionization	$K(4p) + K(nl) + hv \rightarrow K(4s) + K^+ + e^-$	$1.2 \times 10^{-43} \text{ cm}^4 \cdot \text{sec}$	16
Two-photo ionization	$K(4p) + 2hv \rightarrow K^+ + e^-$	$2.9 \times 10^{-49} \text{ cm}^4 \cdot \text{sec}$	16
Energy Pooling	$K(4p) + K(4p) \rightarrow K(4s) + K(5p)$ $K(4p) + K(4p) \rightarrow K(4s) + K(6s)$ $K(4p) + K(4p) \rightarrow K(4s) + K(4d)$	$6.2 \times 10^{-16} \text{ cm}^2$ $1.3 \times 10^{-16} \text{ cm}^2$ $3.1 \times 10^{-16} \text{ cm}^2$	20
Associative ionization	$K(4p) + K(4p) \rightarrow K_2^+ + e^-$	$1.27 \times 10^{-17} \text{ cm}^2$	1
Hornbeck-Molnar	$K(5D) + K(4s) \rightarrow K_2^+ + e^-$ $K(7s) + K(4s) \rightarrow K_2^+ + e^-$	$2.56 \times 10^{-15} \text{ cm}^2$ $2.2 \times 10^{-15} \text{ cm}^2$	21

Table (2): shows photoionization cross sections for potassium atom.

Level	Cross section (Mb)	Reference
4p	7.2	19
5s	0.27	22
3d	28.3	22
5d	28.9	19
7s	0.61	19
6p	3.59 ± 0.18	8
7p	2.21 ± 0.33	8

4. Conclusion

A computational study is carried out to demonstrate the influence of collisional excitation and ionization processes on the temporal evolution of the electron energy distribution function in pulsed laser, excited potassium vapor. Moreover, the evolution of the excited formed states, as well as the generated atomic and molecular ions, are also investigated. The

variation of these parameters with time, laser energy, and potassium vapor density showed nonlinear behavior in the obtained energy spectra of electrons created during the interaction for different values of both laser intensity and potassium vapor density. This nonlinear behavior results from superelastic collisions of the generated free electrons produced through collisional and photoionization processes and the

formed excited states. The computations showed a comparative contribution of Associative and Hornbeck Molnar ionization processes to the production of molecular ions. In general, it is shown that for a reasonable amount of laser energy tuned to the first resonance state of potassium a rapid ionization is possible through photoionization and collisional ionization processes leading eventually to the formation of potassium plasmas of high density in few nanosecond of the laser pulse.

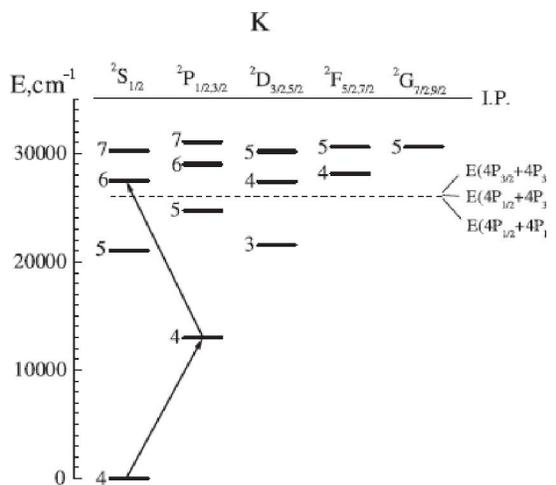


Figure (1) The energy level diagram of potassium atom.

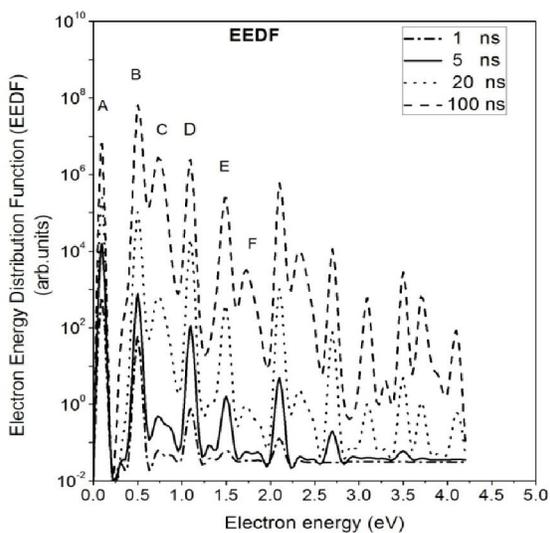


Figure (2): Time development of the electron energy distribution function in potassium vapor excited with laser energy 0.1mJ and potassium vapor density $3.5 \times 10^{15} \text{ cm}^{-3}$.

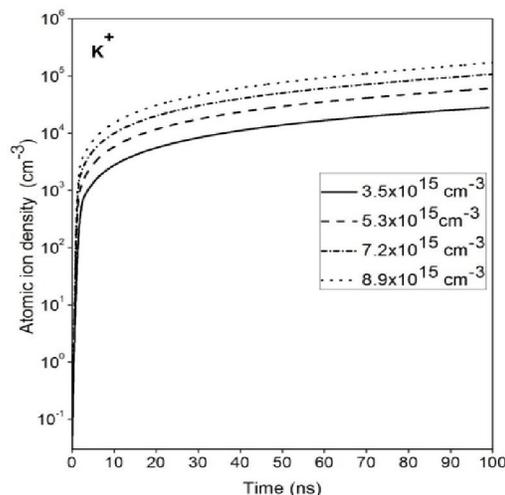


Figure (3): Time evolution of potassium atomic ions (K^+) at different densities and laser energy 0.1mJ.

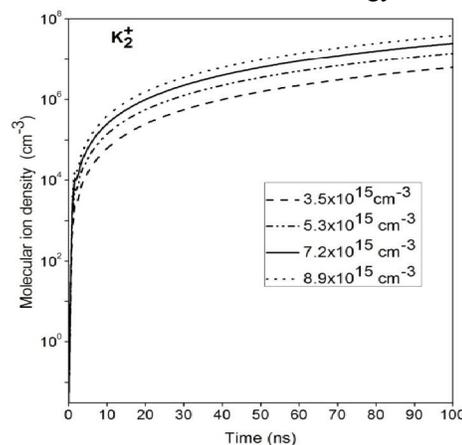


Figure (4): Time evolution of molecular ions K_2^+ at different vapor densities with laser energy = 0.1 mJ.

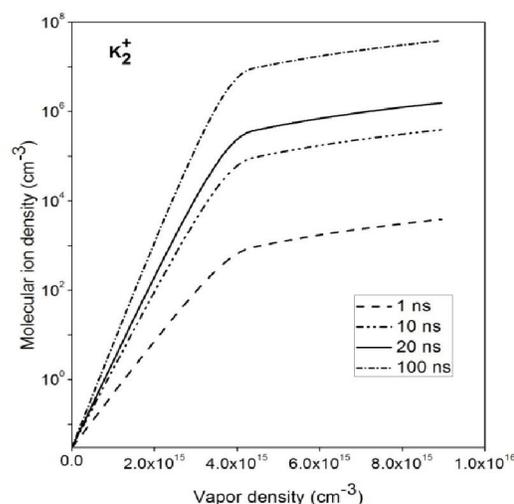


Figure (5): Variation of molecular ions (K_2^+) with potassium vapor density at different times and laser energy = 0.1 mJ.

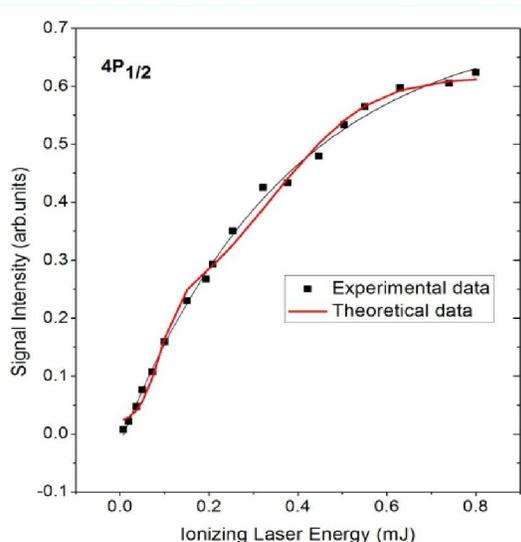


Figure (6): A comparison between the calculated signal intensity with ionizing laser energy (solid line) at 6 ns and the experimentally measured (squares) for the K ($4p\ ^2P_{1/2}$) state.

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