International Journal of New Horizons in Physics

http://dx.doi.org/10.18576/ijnhp/030205

Study of Electron Energy Distribution Function (EEDF) in Potassium Vapor Excited By Nanosecond Laser Pulses

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Received: 21 Feb. 2016, Revised: 22 Mar. 2016, Accepted: 24 Mar. 2016.
Published online: 1 Jul. 2016.

Abstract: A theoretical model of laser excited potassium vapour induced by nanosecond laser pulses tuned to the first resonance, 4s-4p and 4p-nl transitions has been studied. All the important collisional ionization, photoionization, electron collisions and radiative interactions are included in this mode. The numerical model studies the temporal variation of the Electron Energy Distribution Function (EEDF); the dependence of the (EEDF) on potassium vapor density and laser energy; the variation of atomic ion density; molecular ion and electron density. The applied computer simulation model is based on the numerical solution of a set of rate equations that describe the rate of change of the ground and excited states population, beside the atomic ion K^+ density; molecular ion K_2^+ and electron density formed during the interaction. The calculations showed that, the non-equilibrium shape of the electrons occurs due to the relaxation of fast electrons produced by super elastic collision with residual excited potassium atoms. Also the results showed a competition between the processes reponsible for producing K^+. Finally, the model may be useful for interesting applications such as laser isotopes separation and alkali laser systems.

Keywords: Plasma, laser, collisional ionization, photoionization, Rate equations, Alkali metals vapour

1 Introduction

Since the invention of lasers which have found their way into nearly every technology, many scientific, military, medical and commercial laser applications have been developed. One of these fields is studying concerning laser excitation and ionization of alkali-metal vapors, which has been developed in the last decades and becomes the subject of numerous studies nowadays. 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 plasma [1-7]. A better knowledge of collisional and ionization processes in medium containing ground-state atoms, excited atoms and ions was obtained through studying the phenomenon of laser produced plasma[8]. When a gaseous medium is exposed to light with wavelengths equal to resonant transitions in the corresponding atoms, this led to the discovery of photo resonant plasma. Fluorescence from levels lying near twice the excitation energy can be observed. Such fluorescence result from energy-pooling (EP) collisions. Electron Energy distribution function has been exploited in many of plasma sources because it plays an important role in the formation, evolution and radiative properties, so it is considered interested in the plasma physics, fusion and astrophysical communities. In addition to that many applications used the measurements of the (EEDF) such as studies of absolute negative conductivity in photo plasma and light-to-electric signal conversion [9, 10]. Theoretical modeling of alkali metal vapor ionization under resonant optical excitation is a promising field, so it has been studied recently years [11-16]. It is obvious that there are a competition between the ionization processes as single and two photoionization, electron impact ionization, Associative ionization, Hornbeck Molnar ionization, Penning and induced Penning ionization. The aim of this work is to study the variation of the Electron Energy Distribution Function (EEDF) in potassium vapour excitation tuned to the first resonance, 4s-4p with the conditions of (potassium vapour density, laser energy, irradiation time) for the excitation processes as it is important in ionization-guiding discharge experiments.

2 The Theoretical Model of Laser Excited Potassium Vapour 4s→4p Transition

The theoretical model we shall develop is particularly appropriate with low-lying resonance states, such as in the alkali metals Na, K, Rb, etc. Here we consider the case of a homogenous, single constituent, atomic vapor medium

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(potassium vapor) which is irradiated by pulsed laser radiation so that it is tuned to one of the electronic resonance transition.

2.1 Processes considered

As a result of illumination of potassium vapour with a tuned pulsed laser source, the following processes may take place.

1) Two-photon ionization of the 4p level

$$K(4p) + 2h\upsilon_{767.6} \rightarrow K^+ + e^-$$

2) The Penning Ionization

$$K(4p) + K(nl) \rightarrow K(4s) + K^+ + e^-$$

3) The laser induced penning ionization

$$K(4p) + K(4p) + h\upsilon_{767.6} \rightarrow K(4s) + K^+ + e^-$$

4) Electron impact ionization

$$K(nl) + e^-(\varepsilon) \rightarrow K^+ + e^-(\varepsilon_1) + e^-(\varepsilon_2)$$

5) Three-body recombination

$$K^+ + e^-(\varepsilon_1) + e^-(\varepsilon_2) \rightarrow K(nl) + e^-(\varepsilon)$$

6) Radiative recombination

$$K^+ + e^-(\varepsilon) \rightarrow K(nl) + h\upsilon$$

Where 4s ≤ nl≤ 7f

7) Associative ionization

$$K(4p) + K(4p) \rightarrow K_2^+ + e^-$$

8) Hornbeck-Molnar ionization

$$K(5D) + K(4s) \rightarrow K_2^+ + e^-$$
$$K(7s) + K(4s) \rightarrow K_2^+ + e^-$$

9) Super elastic collision (SEC)

$$K(4p) + e^-(\varepsilon_0) \rightarrow K(4s) + e^-(\varepsilon) \qquad where\ \varepsilon > \varepsilon_0$$

10) Electron impact excitation

$$K(nl) + e^-(\varepsilon) \rightarrow K(n'l') + e^-(\varepsilon_0) \qquad where\ \varepsilon > \varepsilon_0$$

11) Absorption of resonant photon

$$K(4s) + h\upsilon_{767.6} \rightarrow K(4p)$$

12) Energy pooling collision

$$K(4p) + K(4p) \rightarrow K(4s) + K(nl)$$

nl= 5p, 6s, 4d.

2.3 Rate equation

Our theoretical model is based on a set of rate equations which are similar to those presented in our previous papers [17]. The rate of change of the population density of the various levels plus for the atomic and molecular ions is given as follows.

The rate of change of the population density of the ground level (4s), (4p) and nl , where n>4p .

$$\begin{aligned}\frac{dN(4s)}{dt} &= N(4p)(R_{21} + A_{21}) - N(4s)R_{12} \\ &\quad + N(4p)\int n_e(\varepsilon)k_{21}(\varepsilon)\,d\varepsilon \\ &\quad - N(4s)\int n_e(\varepsilon)k_{12}(\varepsilon)d\varepsilon \\ &\quad + N(4p)N(n)k_{PI} + \frac{1}{2}N^2(4p)k_{EP} \\ &\quad - N(n)N(4s)k_{HMI} \\ &\quad - N(4s)\int n_e(\varepsilon)k_{1C}(\varepsilon)d\varepsilon \\ &\quad + \frac{1}{2}N^2(4p)\sigma_{pl}\nu F \\ &\quad + N_{k^+}n_e(\varepsilon)[n_e(\varepsilon)k_{c1}(\varepsilon) \\ &\quad + k_{RD}(\varepsilon)] \end{aligned} \qquad (1)$$

$$\begin{aligned}\frac{dN(4p)}{dt} \\ &= N(4s)R_{12} - N(4p)(R_{21} + A_{21}) \\ &- N(4p)\int n_e(\varepsilon)k_{21}(\varepsilon)d\varepsilon + N(4s)\int n_e(\varepsilon)k_{12}(\varepsilon)d\varepsilon \\ &- \frac{1}{2}N^2(4p)k_{AI} - N(4p)N(n)k_{PI} - \frac{1}{2}N^2(4p)\sigma_{pl}\nu F \\ &- \frac{1}{2}N^2(4p)k_{Ep} - N(4p)\sigma_{2C}^{(2)}F^2 \\ &- N(4p)\int n_e(\varepsilon)K_{2C}(\varepsilon)d\varepsilon \\ &+ N_{k^+}n_e(\varepsilon)[n_e(\varepsilon)k_{c2}(\varepsilon) \\ &+ k_{RD}(\varepsilon)] \end{aligned} \qquad (2)$$

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

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

$$\begin{aligned}\frac{dN(K^+)}{dt} &= \sum_{n} N(4p)\,N(n)k_{PI} + \sum_{n} n_e(\varepsilon)\,N(n)k_{nc}(\varepsilon) \\ &\quad + N(4p)\sigma_{2c}^{(2)}F^2 + \sum_{n>2} N(n)\sigma_{nc}^{(1)}F \\ &\quad + \frac{1}{2}N^2(4p)\sigma_{PL}\nu\,F \\ &\quad - N_{k^+}n_e(\varepsilon)\sum_{n}[n_e(\varepsilon)k_{cn}(\varepsilon) \\ &\quad + k_{RD}(\varepsilon)] \end{aligned} \qquad (4)$$

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

Electron energy distribution function equation, which includes all the collisional processes responsible for electrons production in the plasma is given by:

$$\frac{dn_e(\varepsilon)}{dt} = \sum_{n>m} n_e(\varepsilon)\,N(m)k_{nm}(\varepsilon) - \sum_{m<n} n_e(\varepsilon)\,N(n)k_{mn}(\varepsilon) + \sum_n n_e(\varepsilon)\,N(n)k_{nc}(\varepsilon) + \sum_n N(4p)\,N(n)k_{PI} + N(4p)\sigma_{2c}^{(2)}F^2 + \sum_{n>2} N(n)\sigma_{nc}^{(1)}\,F + \frac{1}{2}N^2(4p)\sigma_{PI}\nu F + \frac{1}{2}N^2(4p)k_{AI} + \sum_n N(n)\,N(4s)k_{HMI} - N_{k^+}\,n_e(\varepsilon)\sum_n [n_e(\varepsilon)k_{cn}(\varepsilon) + k_{RD}(\varepsilon)] \quad (6)$$

The normalization conditions are

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

$$\int_0^\infty n_e(\varepsilon)\varepsilon^{1l2}d\varepsilon = 1, \int_0^\infty n_e(\varepsilon)d\varepsilon = N_e \quad (8)$$

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}).
- N_{K^+} represents the density of atomic ions (cm^{-3}). N_{K2^+}
- $N_{K_2^+}$ represents the density of molecular ions (cm^{-3}).
- N_e is the density of free electrons (cm^{-3}).
- No is the density of K atomic vapor (cm^{-3}).
- $n_e(\varepsilon)$ represents the density of free electrons with an energy ε and ε+Δε (cm^{-3}).
- R_{12} is the stimulated absorption rate coefficient from 4s to 4p level (se^{c-1}).
- R_{21} is the stimulated emission rate coefficient from 4p to 4s level (sec^{-1}).
- A_{nm} Einstein coefficient for spontaneous emission for the transition n→m (sec^{-1}).
- k_{21} represents the electron collision rate coefficient for transition from (4p→4s) as a function of electron energy (ε) ($cm^3.sec^{-1}$).
- k_{12} represents the electron collision rate coefficient for transition from (4s→4p) as a function of electron energy (ε) ($cm^3.sec^{-1}$).
- $\sigma_{nc}^{(1)}$ represents the single photon ionization cross-section (cm^2) [18, 19].
- $\sigma_{2c}^{(2)}$ represents the two photon ionization cross-section ($cm^4\ J^{-2}\ sec$)= 2.9×10^{-49} $cm^4.sec$ [20].
- F photon flux (photon $cm^{-2}\ sec^{-1}$).
- υ represents the relative velocity of the free electrons ($cm\ sec^{-1}$).
- k_{1c} represents electron impact ionization rate coefficient for level 4s ($cm^3.sec^{-1}$).
- k_{2c} represents electron impact ionization rate coefficient for level 4p ($cm^3.sec^{-1}$).
- $k_{nc}(\varepsilon)$ represents electron impact ionization rate coefficient for level n ($cm^3.sec^{-1}$) [21].
- k_{cn} represents the three body recombination rate coefficient for n level ($cm^6.sec^{-1}$) [21].
- k_{nm} represents the electron collision rate coefficient for transition from (n→m) as a function of electron energy (ε) ($cm^3.sec^{-1}$)
- $k_{mn}(\varepsilon)$ represents the electron collision rate coefficient for transition from (m→n) as a function of electron energy (ε) ($cm^3.sec^{-1}$).
- k_{RD} represents the radiative recombination rate coefficient ($cm^3.sec^{-1}$)[21].
- k_{PI} represents Penning ionization rate coefficient for level n ($cm^3.sec^{-1}$).
- k_{EP} represents the energy pooling collisions rate coefficient ($cm^3.sec^{-1}$) [22].
- k_{AI} represents the association ionization rate coefficient ($cm^3.sec^{-1}$)= 1.27×10^{-17} cm^2 [23].
- k_{HMI} represents the Horn-beck Molnar ionization rate coefficient ($cm^3.sec^{-1}$) [24].
- σ_{PI} represents Penning ionization cross section (cm^2).
- σ_{pl} represents induced Penning ionization cross section (cm^2) =1.2×10^{-43} $cm^4.sec$ [20] .

Note that the factor $1/2$ with k_{EP} and k_{AI} corrects for possible double counting of each colliding pair of identical particles [25].

3 Results and Discussion

Equations from (1) to (8) are solved numerically using the Rung-Kutta fourth-order technique under the experimental conditions of Amin et al [18]. In their experiment, they used dye-laser beam, the laser energy varied from 0.1 mJ to 1mJ, using potassium vapor density varying in a range from

($3.57 x 10^{15}$ to $8.94x10^{15}$ cm^{-3}) and pulse duration ≈6 ns. A computer program is undertaken to obtain the following relations:

1) Variation of electron energy distribution function with time, laser energy, potassium vapor density.
2) Variation of electrons density (n_e), atomic (K^+) and molecular ions (K_2^+) density with potassium vapor density.
3) Comparative study of the ionization processes produced atomic and molecular ions.

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 (1). These calculations enable us to study the time evolution of the electrons seeding and growing processes. From this figure we note that the spectral structure can be seen from the noticeable increase of the height of the peaks (A, B, C, D, E and F) lying at energies 0.08, 0.5, 0.7, 1.07, 1.49 eV respectively. These peaks are attributed to some physical processes which contribute to the population and depopulation of the various excited states considered in this analysis. The energy distribution of the electrons produced in peak A which is centered at a mean energy ε ≈ 0.08 eV created by Horn-beck Molnar ionization of 7s and 5d and single photo ionization of 3d and 5s, which are described by the following equations:

$$K(5d,7s) + K(4s) \rightarrow K_2^+ + e^-$$

$$K(3d,5s) + h\nu \rightarrow K^+ + e^-$$

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

$$K(4p) + K(4p) + h\nu \rightarrow K(4s) + K^+ + e^-$$

$$K(4p) + 2h\nu \rightarrow K^+ + e^-$$

$$K(4p) + K(4p) \rightarrow K_2^+ + e^-$$

While the peak C corresponds to single photo ionization of 6p, which are described by the following equations:

$$K(6p) + h\nu \rightarrow K^+ + e^-$$

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

$$K(\mathrm{4p,5d,7s,7p}) + h\nu \rightarrow K^+ + e^-$$

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

$$K(5p) + e^- \rightarrow K^+ + 2e^-$$

We can say that, the others peaks are attributed to super elastic collision which is described by the following equations:

$$K(4p) + e^-(low\ energy) \rightarrow K(4s) + e^-(high\ energy)$$

At this point, we can conclude that the ionization of K atom based on laser saturation is mainly produced through single, two photoionization, 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 super elastic collisions are responsible for the heating of electrons resulting on rapid ionization of the medium [22].

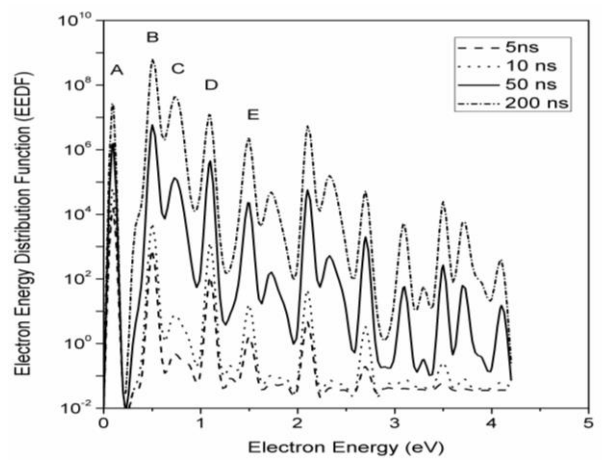

Figure (1): The spectral structure of the electron energy distribution function as a function of time in potassium vapor excited with laser energy 0.1mJ and vapor density $3.5x10^{15}$ cm^{-3}.

3.2 Variation of electron energy distribution function with laser energy.

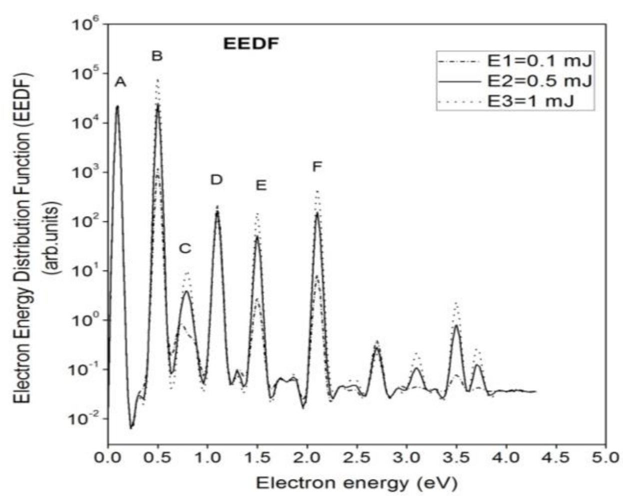

Figure (2): Varaiation of the electron energy distribution function with laser energy at 6 ns from the laser pulse and potassium vapor density equal to $3.5x10^{15}$ cm^{-3}.

The electron energy distribution function at different laser power density is shown in figure (2). From this figure we show that, electron energy distribution function increases as the laser power increases. Approximately the same results are obtained in peaks A, B, C, D, E, F, this result indicates the dependence of the occurrence of some physical processes on the laser power which in turn results in the appearance of such peaks in EEDF.

3.3 Variation of electron energy distribution function with potassium vapor density

The dependence of the electron energy distribution function on the potassium vapor density at 6ns and laser energy = 0.1 mJ is indicated in figure (3). This figure enables us to study the evolution of the electrons seeding and growing processes. Also this figure confirmed the observed peaks (A, B, C, D, E and F) shown in figure (1) which increased in height as potassium vapor density increases. However the interesting point from our point of view is that, the collisional ionization and photo ionization are the dominant processes during the plasma formation in laser excited potassium vapor [26-29].

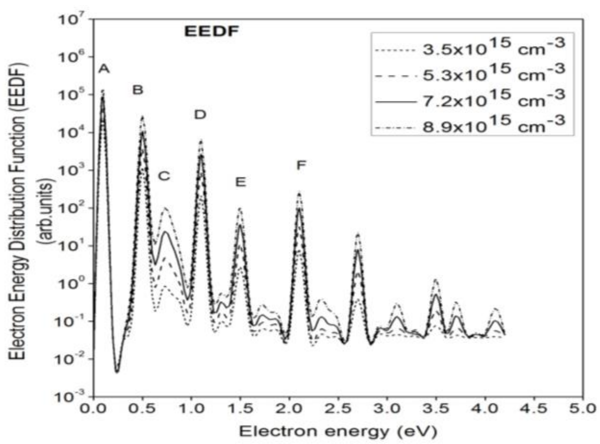

Figure (3): Electron Energy Distribution Function after 6 ns from the laser pulse at different potassium vapor densities and laser energy 0.1mJ.

3.4 Variation of electrons density (n_e), atomic (K^+) and molecular ions (K^+_2) density with potassium vapor density

In addition, we have studied the variation of electrons density (n_e), atomic (K^+) and molecular ions (K^+_2) density with potassium vapor density at 10 ns and laser energy equals to 0.1mJ as shown in figure (4). The rate of production of molecular ions K^+_2 is greater than the atomic ions K^+. This confirms that Horn-beck Molnar and Associative ionization for producing the molecular ions are more effective than single and two photo ionization, Penning and induced Penning ionization for producing the atomic ions.

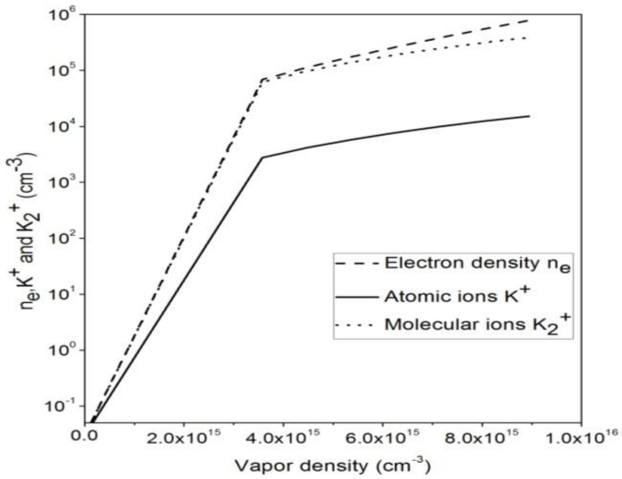

Figure (4): Variation of electron density (n_e), atomic and molecular ions (K^+, K^+_2) with potassium vapor density at time 10 ns and laser energy = 0.1 mJ.

3.5 Comparative study of the ionization processes producing atomic ions.

The rate of atomic ions density which is produced by Penning ionization, induced Penning ionization, two photoionization (of all 4p atoms) and single photo ionization (of all excited states) at potassium vapor density = 3.5x10^{15} cm^{-3} and laser energy = 0.1 mJ is shown in figure (5). This figure shows that the two photoionization is the most effective process in the production of atomic ions up to 0.1 ns then the induced Penning ionization overcome. We note that Penning, induced Penning ionization and two photoionization are increasing with time. As shown in the following equations.

$$K(4p) + K(4p) \rightarrow K(4s) + K^+ + e^- \quad \text{(Penning ionization)}$$

$$K(4p) + K(4p) + h\nu \rightarrow K(4s) + K^+ + e^-(\text{ induced Penning ionization})$$

$$K(4p) + 2h\nu \rightarrow K^+ + e^-(\text{Two photoionization})$$

However, single photoionization remains constant until 0.02 ns then increased with time. This can be interpreted as in the times less than 0.02 ns, the excited states not filled yet. Also a comparison between the density of atomic ions which produced by Penning ionization, induced Penning ionization, single photoionization and two photoionization with laser energy variation are shown in figures (6).

The figures clarified that the contribution of the induced Penning ionization and two photoionization are greater than the other two processes in atomic ions production. From the two figures it is clear that the density of atomic ions (which produced by each process) increases with the increase of both potassium vapor density and laser energy. This result reveals that, the major processes for plasma creation in potassium vapor are the induced Penning ionization and two photoionization processes.

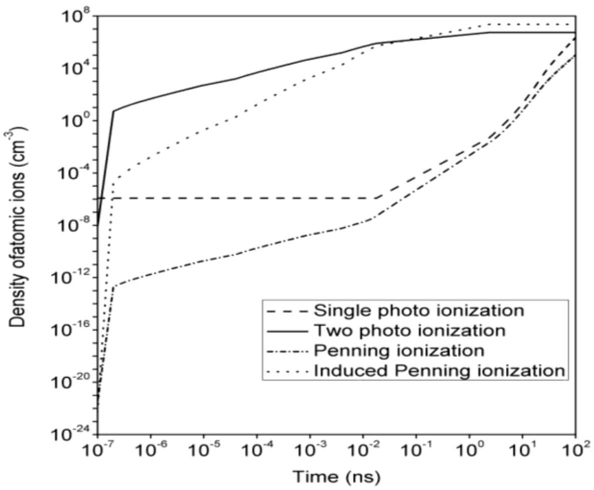

Figure (5): Rate of atomic ions production by Penning, induced Penning, single and two photoionization (at potassium vapor density = $3.5x10^{15}$ cm^{-3} and laser energy = 0.1 mJ).

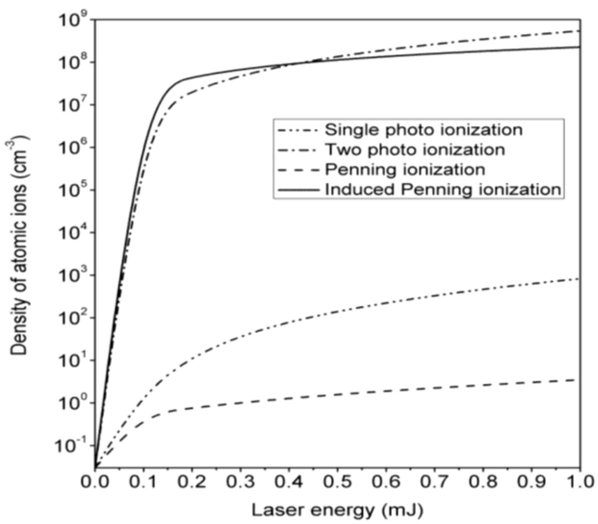

Figure (6): comparison between atomic ions, which produced by Penning, induced Penning, single and two photoionization with laser energy variation (at 6 ns and potassium vapor density=$3.5x10^{15}cm^{-3}$).

4 Conclusion

A computational study has been carried out on the influence of collisional excitation and ionization processes on the electron energy distribution function as well as the density of atomic, molecular ions and electrons in pulsed laser excited potassium vapour. The results showed a nonlinear behavior of the energy spectra of the electrons created during the interaction for different values of both the laser intensity and the potassium vapour density. This non-equilibrium shape results from super elastic collisions between the generated free electrons resulting from collisional ionization and photoionization with the formed excited states. The calculations also showed a competition exists between the collisional ionization (Penning ionization and induced-Penning ionization) and photoionization processes (single and two photoionization) in producing atomic ions. From our model we could study the variation of electrons density (n_e), atomic (K^+) and molecular ions ($K^+{}_2$) density with potassium vapor density. The model may be useful in a laser induced plasma or discharge experiments.

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