Excitation and ionization of laser-pumped potassium vapour

M A Mahmoud

Physics Department, South Valley University,
Faculty of Science, Sohg - Egypt

Received 30 June 2003; revised 20 November 2003; accepted 1 March 2004

The excitation and ionization of low dense K vapour \((n \approx 10^{14} \text{ cm}^{-3})\) by resonant cw laser radiation \((I \approx 10^9 \text{ w. cm}^{-2})\) have been studied theoretically and the processes responsible for the transfer of energy from the laser - excited atoms into ionization have been presented. It has been established that seed electrons created by collisional ionization processes were heated in superelastic collisions with laser excited atoms, and subsequent electron - impact excitation and ionization as well as Penning ionization of high lying levels lead to the creation of more electrons.

IPC Code: H 01S 309

Keywords: Collisional ionization, Superelastic collisions, Electron energy distribution function, Energy pooling collisions

I Introduction

Resonant laser excitation is a very efficient technique for coupling energy into a vapour. In 1976 Lucatorto and McIlrath\(^1\) observed for the first time complete ionization of dense sodium vapour irradiated by high - power pulsed laser. At about the same time, Allegrini \textit{et al.}\(^2\) observed fluorescence from high - lying levels in sodium vapour excited with cw laser tuned to the first resonance transition. The population of these high - lying levels was attributed to the energy pooling collisions. Since then, efficient ionization by resonance radiation has been seen in all alkali -metal and alkaline - earth metal vapours tested [Na(Refs3,4), Li(Ref.5), Rb(Refs 6-8), K(9-11), Cs(12,13), Ba(14,15), Sr and Ca(Ref.16)].

A theoretical model was suggested by Measures\(^17\) in 1970 to explain laser ionization based on resonance saturation (LIBORS). A computer code\(^18-20\) was subsequently developed to simulate the experiments that carried out by Lucatorto and McIlrath\(^1\). The model assumes that laser saturates the resonance transition and initially provides seed electrons by numerous processes best summarized by Kopystynska and Moir\(^21\). These ionization mechanisms include, for example, two-photon ionization from 3p state and associative ionization between two excited sodium atoms. The model then predicts that the seed electrons gain kinetic energy by depopulating the 3p level through superelastic collisions (SEC). These hot electrons then collisionally ionize electrons from 3p level or excite them to higher levels from which they can photoionized. Avalanche ionization should then ensue with the energy necessary for ionization being supplied by continually replenishing the 3p population.

On the other hand, a theoretical model developed by Morgan\(^22\) to incorporate a more realistic non - Maxwellian electron energy distribution during the early stages of ionization assumes that collisional ionization is the dominant process in a laser excited sodium plasma.

Most of the calculations of the theoretical studies for the ionization of metallic vapours, which are based on resonance excited by laser have used different approaches, (i) the assumption of a Maxwellian distribution which was suggested by Measures \textit{et al.}\(^18,19\), i.e \(f(E) \approx E^{3/2} \exp(-E/kT_e)\). Therefore the only quantity which needs to be known in this case is the electron temperature \((T_e)\). (ii) even when a non - Maxwellian distribution is presented by Morgan\(^20\). Only an approximate value for the rates was used which in turn
could only give a tentative investigation for the exact electron energy distribution function.

In this work a calculation model which previously developed and applied successfully for the case of sodium, will be briefly described to study the kinetics of excitations and ionization processes in laser pumped potassium vapour. The model focuses on the time-dependent electron energy distribution function as well as a set of rate equations describing the temporal behaviour of the excited states, atomic and molecular ions.

2 Formulation of the Numerical Model

The numerical procedure of laser - pumped potassium (K) vapour which is described as follows:

2.1 Rate coefficients

The computational model consists of a large number of rate equations describing about 15 excited states of K atom, the atomic and molecular ions species as well as the energy distribution of the free electrons created during the interaction. The model includes various elementary physical processes such as associative ionization, energy pooling collisions, inelastic and superelastic collisions, etc. These various physical processes are shown in Table 1.

The rate coefficients of these various physical processes are taken from analytical expression given in Refs (25, 26) with some approximations made to fit our calculations. Due to the lack of data concerning the rate coefficients of Penning ionization for the case of potassium as an approximation we followed the same consideration used by Huuennkens et al. to take the value of this coefficient as measured for Rb (Refs 6-8).

Under the present experimental conditions (the laser power density $\leq 10^7$ wcmt$^2$) photo-ionization processes from either ground or excited states do not play an important role. Therefore it has been ignored in our calculations.

2.2 Formulations

Considerable physical insight into various physical processes considered in our model during the interaction of laser radiation with K vapour leading to its saturation through a resonance transition can be gained in Fig. 1. We have formulated a computer analysis that utilizes the 15-level model of the K atom as shown in Fig. 1.

In these calculations we considered the fact that, under laser saturation conditions the redistribution of the population between the two laser coupled states in a time is approximately given by:

$$\tau_v = \frac{1}{(1 + g) R_{21}}$$  \hspace{1cm} (1)$$

where $g = g_2 / g_1$ the ratio of the degeneracy of the upper and lower levels, and

$$R_{21} = \frac{1}{4\pi} \int B_{21}(v) L_{21}(v) dv = \frac{l'(v)}{h v} \sigma_{21} \ldots$$  \hspace{1cm} (2)$$

$$\sigma_{21} = \frac{h v}{4\pi} \int B_{21} L_{21}(v) dv \ldots$$  \hspace{1cm} (3)$$

$R_{21}$ represents the stimulated emission rate and $B_{21}$ is the Milne stimulated coefficient for the resonance transition. $\sigma_{21}$ represents the frequency integrated stimulated emission cross section, $l'(v)$ the laser spectral irradiance, and $L_{21}(v)$ represents the resonance line profile function.

The saturation condition can be expressed in the form $l'(v) = I_1(v)$, then the saturated spectral irradiance

$$I_1(v) = \frac{h v}{\sigma_{21}} \tau_{21} \tau_{21} \tau_{21}^\text{RAD} \ldots$$  \hspace{1cm} (4)$$

Where $\tau_{21}^\text{RAD} = 1 / A_{21}$ \hspace{1cm} (5)$$

The ratio of the resonance to the ground state population densities closely approximates the infinite temperature limit viz,

$$\frac{N_2}{N_1} \equiv \frac{g_2}{g_1} = g \ldots$$  \hspace{1cm} (6)$$

Under the saturation conditions, the general rate equation that describes the temporal behaviour of the population density of levels 4s is expressed as:

$$\frac{dN(4s)}{dt} = N(4p)(R_{21} + A_{21}) - N(4s)R_{12} + N_1(\varepsilon)N(4p)K_{21}(\varepsilon) - N_1(\varepsilon)N(4s)K_{12}(\varepsilon) + N(4p)K_p + N^0(4p)K_e \ldots$$  \hspace{1cm} (7)$$

where $N(4s), N(4p)$ and $N(n)$ represent the population densities of levels 4s, 4p and n respectively. $N_1(\varepsilon)$ is the free electrons density as a function of electron energy $\varepsilon$. $K_p (\text{cm}^3 \text{sec}^{-1}), K_e (\text{cm}^3 \text{sec}^{-1})$ and $K_e (\text{cm}^3 \text{sec}^{-1})$ are the rate coefficients of associative ionization, Penning ionization and energy pooling collisions, respectively. $A_{21} (\text{sec}^{-1})$ represents the Einstein coefficient for the transition 2 to 1. $K_{21}(\varepsilon)$ represents the electron
Table 1—Kinetic processes in laser excited potassium vapour

<table>
<thead>
<tr>
<th>Process</th>
<th>Rate</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>K(4p) + K(4p) → K(4s) + e</td>
<td>(K_e = 2.7 \times 10^{11} \text{ cm}^3 \text{ sec}^{-1})</td>
<td>10</td>
</tr>
<tr>
<td>K(4p) + K(4p) → K(6s) + K(4s)</td>
<td>(K_p = 5.4 \times 10^{11} \text{ cm}^3 \text{ sec}^{-1})</td>
<td>11</td>
</tr>
<tr>
<td>K(4p) + K(4p) → K(4d) + K(4s)</td>
<td>(K_p = 7.2 \times 10^{11} \text{ cm}^3 \text{ sec}^{-1})</td>
<td>11</td>
</tr>
<tr>
<td>K(4p) + K(nl) → K + K(4s) + e</td>
<td>(K_n = 10^7 \text{ cm}^3 \text{ sec}^{-1})</td>
<td>7</td>
</tr>
</tbody>
</table>

where \(K\_e (\text{cm}^3 \text{ sec}^{-1})\) represents the electron impact ionization rate coefficients.

For any higher excited state \(nl\) the rate equation is represented by:

\[
\frac{dN(n)}{dt} = \sum_{m>n} N\_e(\epsilon)N(n)K\_nm(\epsilon) - \sum_{m<n} N\_e(\epsilon)N(m)K\_mn(\epsilon) \sum_{n} N(n)K\_nc(\epsilon) - N(4p)\sum_{n} N(n)K\_pf + N^2(4p)K\_ep + N^2(4p)[N\_e(\epsilon)K\_nc(\epsilon) + K\_rd(\epsilon)] \]

where \(N(m) (\text{cm}^{-3})\) represents the population density of level \(m\), \(N\_e (\text{cm}^{-3})\) is the free electron density, \(A\_mn (\text{sec}^{-1})\) represents the Einstein spontaneous transition probability for levels \(n\) to \(m\), \(K\_nm (\text{cm}^3 \text{ sec}^{-1})\) represents the electron – collision rate coefficient for transition \(m \rightarrow n\), \(K\_nc (\text{cm}^3 \text{ sec}^{-1})\) and \(K\_nm (\text{cm}^3 \text{ sec}^{-1})\) are the rate coefficients of the three body recombination and the radiative recombination respectively.

The time evolution of the electron energy distribution function is given by Boltzmann equation which includes most of the collisional processes as given in Table 1 to be as:

\[
\frac{dN\_e(\epsilon)}{dt} = \sum_{m>n} N\_e(\epsilon)N(m)K\_nm(\epsilon) - \sum_{m<n} N\_e(\epsilon)N(m)K\_mn(\epsilon) \sum_{n} N(n)K\_nc(\epsilon) - N(4p)\sum_{n} N(n)K\_pf + N^2(4p)K\_ep + N^2(4p)[N\_e(\epsilon)K\_nc(\epsilon) + K\_rd(\epsilon)]
\]

- collision rate coefficient for transition 2 to 1.
2.3 Method of calculation
In these calculations the electron energy ($E$) lies between 0 to 6 eV (> the ionization energy of potassium atom). Considering the energy step order 0.1 eV, therefore about 60 rate equations representing the growth rate of the electron energy distribution function have to be calculated each time step. This, in addition to the set of the rate equations describing the time evolution of the excited states of the population density as well as the ionic specie formed during the interaction. These equations are solved numerically using a Runge technique, taking into account the rate coefficients for the various processes given in Table 1.

3 Results and Discussions
The computer program was run to calculate firstly the time evolution of the electron energy distribution function under the experimental conditions of Allegrini et al. In their experiment the potassium density was taken as $1.2 \times 10^{14}$ cm$^{-3}$ and the cw laser irradiance was $10^4$ W/cm$^2$.

In Fig. (2) the calculated electron energy distribution function is shown at various time intervals namely 1, 20, 100 and 500 ns respectively. From Fig. 2 we notice that the spectral structure is observed with certain peaks (a, b, c, d, e and f) lying at energies 0.1, 0.8, 1.6, 2.4, 3.2 and 4.5 eV respectively. Inspect these curves we attributed the peak labeled a to associative ionization process. Since in this reaction the relative kinetic energy of the electron is typically much less than 0.1 eV (the difference of the binding energy of the molecular ion and the ionization energy of the K(4p) state. On the other hand the peak b in the same figure attributed to the electrons created by Penning ionization process. The peaks c, d, e and f created by electrons initiated from associative ionization and Penning ionization and heated up through superelastic collisions. These peaks are even more clearer as the time increases which confirm the idea that during the interaction associative ionization; Penning ionization are source of the free electron where as superelastic collisions resulting in an energy gain processes.

Fig. (2) - Time development of the electron energy distribution function in potassium vapour excited with cw laser
At this point we can conclude that the calculated electron energy distribution function at different time intervals confirms the experimental observations of the plasma created under resonant laser excitation. Moreover the population of the highly excited states through inelastic excitations by considerably energetic electrons as well as energy pooling collisions process was confirmed the increase of the peak amplitude with increase of the time intervals.

The calculated values of the population density of two selected highly excited states (i.e K (6s) and K(4d)) are plotted as function of time (Fig. 3). The rate of growth of both the states increases as the time increases. This may be due to the continuous population of these levels through energy pooling collisions process between two up 4p potassium atoms. These two levels 4d, 6s have chosen deliberately because there are measured values for their energy pooling collision rate coefficients (Table 1). The rate of growth of 4d state exceeds that for 6s state (Fig. 3). This confirms the high rate coefficient value for the 4d state observed experimentally.

Similarly, plotting is represented by the variation of the population density of atomic and molecular ions \( (K^+, K^+_{2}) \) as a function of time. These are illustrated in Fig. 4. From Fig. 4 we can also see that, the rate of growth of \( K^+ \) as well as \( K^+_{2} \) increases sharply during the early stages of the plasma formation. The interpretation of this ionic behaviour can be attributed to the fact that, associative ionization and Penning ionization processes, which play an important role during the early stages of the plasma mainly, produce these ions. Therefore, as the time increases these processes become less active which is indicated by this low growth of both ionic species over the interval from 100 - 500 ns. The \( K^+ \) is growing faster than the \( K^+_{2} \) on the time interval from 10 to 300 ns as where the \( K^+_{2} \) starts to grow rapidly reaching a value of the same order of that the \( K^+ \) at the end of time interval 500 ns (Fig. 4). This behaviour indicates the important role of the associative ionization process in producing the seed electrons which initiate the full ionization of atomic vapour. On the other hand the rapid growth of the \( K^+ \) during the late stages of the plasma formation can be
attributed to the rate of growth of inelastic ionization process between the energetic electrons and the neutral atoms leading to their ionization and eventually to the atomic ions formation which in turn increases their population. Besides these atomic ions processes the low values of the K* over the whole time intervals may be attributed to the radiative recombination and three body recombination processes, which act to deplete these K* leading to this, observed low values.

4 Conclusions
We have applied a previously developed model to study the transient kinetic of the ionization of the potassium vapour under saturation conditions with cw laser excitation. The calculations revealed that the electron energy distribution function is highly non-Maxwellian and is characterized by spikes structure over the whole range of the energy considered in our calculations. It has been found that this spikes structure represents the electron spectrum created due to the associative ionization and Penning ionization and heated up through three successive superelastic collisions. Energy pooling collisions play an important role in populating the highly excited states of the potassium vapour.

References