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Analytical Study of The Effect of Loss Processes on Laser Induced Breakdown in Argon Gas

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□ ABSTRACT □

In the present work, a modification of a previously developed electron cascade model is presented. More than one electronic state is incorporated in the model as well as various loss processes. Thus, new inelastic process such as photo ionization and electron impact ionization of the electronic excited state of the argon atom are considered. Moreover, loss processes which may act to deplete the number of electron such as electron diffusion; three body recombination and dissociate recombination are taken into account. All these processes are incorporated into the time-dependent Boltzmann equation which is solved numerically simultaneously with set of rate equations for both the electron energy distribution function (EEDF) and the excited state population respectively.

Key words: Argon laser gas - breakdown in argon gas,

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دراسة تحليلية لتأثير عمليات الفقد (الضياع)
على انهيار غاز الأرغون تحت تأثير أشعة الليزر

الدكتور بسام غزولين*

(تاريخ الإيداع 6 / 11 / 2013. قُبِل للنشر في 15 / 4 /2014)

□ ملخّص □

في هذه الدراسة طورنا، بشكل واضح، نموذج الالكترونات المتعاقبة، فضلاً عن ذلك أكثر من مجموعة سويات طاقية، وكذلك مختلف أنواع عمليات الفقد. وهكذا الخواص الجديدة غير المرنة، مثل: التأين الضوئي والتأين بالتصادم التعاقبي للسويات المثارة لذرات غاز الأرغون المدروسة. إن عمليات الفقد التي من الممكن أن تؤثر في تقليل عدد الالكترونات مثل تبعثر أو تشتت الالكترونات من الحجم البؤري، إعادة التوحيد ثلاثي الجسيمات، وإعادة التكوين التفككي جمعيها أخذت بالاعتبار. وكل هذه الخواص مدمجة بمعادلة بولتزمان المتغيرة مع الزمن، و التي حلت بشكل عددي ومتزامن مع وضع معدل دالة توزيع الإلكترونات وتابع التوزع الالكتروني للحالة المثارة يشكل متعاقب

الكلمات المفتاحية: انهيار ذرات الارغون في الليزر– توزع وانتشار طاقة الالكترونات في ذرات الأرغون.

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Introduction:

The argon atom was chosen in our investigation for two reasons.
First, argon is frequently use rare gas in laboratory studies of plasma and industrial application. Secondly, this gaz pronounced Ramsauer minimum and consequently a collision cross section which is strongly dependent on electron energy.

Laser induced breakdown of gases has been the subject of many experimental and theoretical investigations in a number of review papers [1,2]. However, most of the theoretical works carried out so far, impose some simplifying assumptions. One of the fundamental assumptions in almost all extinctive computation, is the use of the Maxwelluain electron energy distribution function**.** There are two mechanisms that can lead to breakdown in gases under the action of high intensity of laser pulses, namely multiphoton ionization and electron cascade ionization [3,4].

A simple electron are developed [5,6,] cascade model based on the step by step integration of the electron energy distribution function when the electrons gain continuously their energy from the electromagnetic field of laser beam [7,8].When their energies are sufficiently large, they may cause excitation or ionization in inelastic collisions with gas atoms. Hence, further electrons are liberated either by direct collisions between electrons and neutral atoms (or excited atoms), or otherwise through the photoionization of the excited atoms by the laser field it self. In this way an avalanche or cascade of electrons will be developed in the focal region. The continuous electron energy range is approximated by a series of closely spaced steps. Therefore, [7] the rate of the energy gain by free electrons from the laser field is represented by the equation:

$$\partial f/\partial t = G(\varepsilon)(\partial f/\partial \varepsilon) + 2\varepsilon G(\varepsilon)(\partial^2 f/\partial \varepsilon^2) \qquad (1)$$

Where, ε is the electron energy

$$G(\varepsilon) = (1/3)\varepsilon_0 \nu_m(\varepsilon)$$
$$\varepsilon_0 = e^2E^2/2m\omega^2 = (377e^2/2m\omega^2)I(t) \qquad (2)$$

$\varepsilon_:$ is the oscillatory energy of an electron of charge (e) and mass (m) in laser with electric field amplitude (E) and angular frequency(ω).

$f=f(\varepsilon,t)$: is the electron density at any time t , having energy between (ε) and $(\varepsilon+\Delta\varepsilon)$.

$\nu_m(\varepsilon)$: is the momentum transfer collision frequency between electrons and gas atoms.

The first term in equation (1) represents the energy gain, while the second term was referred to by the authors as the energy diffusion term, in analogy to the spatial diffusion of particles. This diffusion term is considered to be very important, especially for rare gases where the first excited state lies relatively high with respect to the ground state. This allows for the possibility of an electron gaining appreciably more (or less) than the mean energy in a collision. Therefore an electron may move from energy less than ε_x to energy larger than ε_i; thus ionization would be more likely.

This equation was solved using a step by step integration to obtain a complete temporal variation of electron energy distribution during the whole flash duration.

An important idea in their model was the ionization of excited atoms by collisions with low energy electrons “two step ionization “. This yields a faster than exponential

growth rate, since this process depends on the product of electrons and excited atoms concentration: and the latter is proportional to the electron concentration also.

Their model was successfully applied to interpret the breakdown phenomena of helium gas irradiated by ruby laser.

The development of the cascade mechanism is considered through two consequent stages. In the early one, electron impact excitation and ionization and multiphoton ionization of the ground state atoms are considered as the processes which cause the cascade development. In addition to these processes the loss due to electron diffusion is also accounted for as an important process. In the latter stage of the cascade, when the density of electrons is high enough , more processes are included, besides those mentioned above, such as the three body recombination, dimer formation, dissociative recombination and photoionztion of excite atomic states formed.

The following processes are responsible for the change of the number of electrons in any energy step (j), during an interval of time (Δt):

1- Electrons can move to the next higher energy step, by absorbing energy from the electromagnetic field of the laser beam during an elastic collision with neutral atoms, inverse bremsstrahlung absorption process, equation (1).

Together with this process, electrons may lose an mount of their energy (2 $m\varepsilon_0 \nu_m / M$), Where M and m are the atomic and electronic masses respectively; and other symbols are as described in equation (1). This elastic process is expressed as follows:

$$A + e(\varepsilon) + h\nu \rightarrow A + e(\varepsilon') \quad , \varepsilon' > \varepsilon \qquad (3)$$

2- Electrons whose energies are higher than the excitation energy (ε_{xL}) of any atomic excited state $L,(L=1,2,.....)$ may lose a part of their energies in exciting a neutral atom , such as:

$$A + e(\varepsilon) \rightarrow A^{*L} + e(\varepsilon - \varepsilon_{XL}) \quad , \varepsilon \geq \varepsilon_{xL} \qquad (4)$$

This process is considered as a loss term of electrons in energy step j and also as a gain term of electrons in energy step $(\varepsilon - \varepsilon_{xL})$

3- Electrons having energy greater than the ionization potential energy (ε_I) of a gas spend a part of their energies in ionizing a ground state atom.

$$A + e(\varepsilon) \rightarrow A^{+} + 2e(\varepsilon - \varepsilon_i) \quad , \quad \varepsilon \geq \varepsilon_i \qquad (5)$$

After each ionization process, a new electron is born and this is assumed to have zero electron energy. Thus, this process is simultaneously considered as a loss term for electron in energy step (j) and a gain term for electrons having energies $(\varepsilon_j - \varepsilon_i)$ as well electrons in energy step ($J= 1$).

4- Electrons can enter energy step ($j=1$) due to ionization of ground state atoms by the atomic absorption of a number k of photons from the laser field (where, k $h\nu \geq \varepsilon_i$)

$$A + kh\nu \rightarrow A^{+} + e \quad , \quad k = < \varepsilon_i / h\nu > \qquad (6)$$

5- Electrons having any energy higher than zero electrons energy can escape out of the focal volume due to diffusion (this process occurs, especially, at low pressures or for small focal volumes).

The aforementioned processes may occur at the early stages of the cascade development. In addition to these, the following processes may take place during the late stages of the cascade development.

6- Electrons with energies higher than the difference between the ionization and excitation energies of any excited state *L*, can ionize an excited atom (A^{*L}) and drops to a lower electrons energy step.

$$A^{*L} + e(\varepsilon) \rightarrow A^{+} + 2e \quad , \quad \varepsilon \geq \varepsilon_i - \varepsilon_{xL} \qquad (7)$$

This process is considered as a loss term for electrons in energy step(j), and also as gain term for electrons in both energy step $[\varepsilon_j - (\varepsilon_i - \varepsilon_{xL})]$ and energy step ($j=1$).

7- Electrons can also enter step ($j= 1$) due to the photoionization of the excited atoms (A^{*L}) by their absorption of photons from the laser field (where, $k_L\, h\nu \geq \varepsilon_i - \varepsilon_{xL}$)

$$A^{*L} + k_L h\nu \rightarrow A^{+} + e \,,\; k_L = \langle (e_i - e_x)/h\nu \rangle \qquad (8)$$

8- Electrons can be lost from step j due to their recombination with an ion through a three body recombination process.

$$A^{+} + e + e \rightarrow A^{*L} + e \qquad (9)$$

9- Electrons can also be lost from step j due to their recombination with dimer ions,

$$A^{*} + e \rightarrow A^{*L} + A \qquad (10)$$

It is obvious that, these various processes may result in changing the concentration of excited atoms in different excited energy states. Therefore it is worthwhile to consider the net rate of change of the excited atoms concentration of any energy state, during an interval of time (Δt).

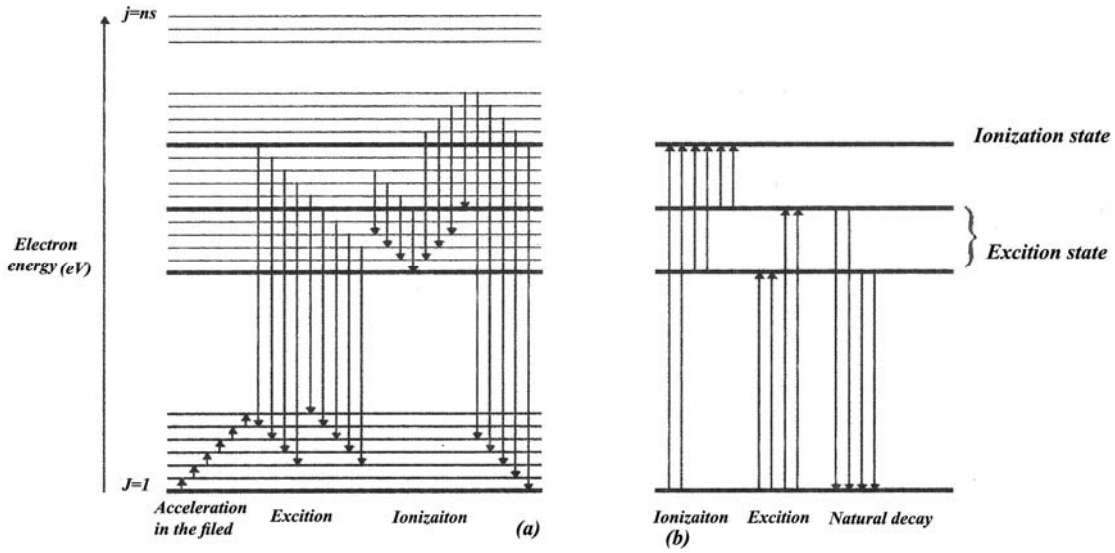


Figure (1) (a) the possible transition of free electrons among the various energy state.
(b) The possible transition of atoms among their atomic energy levels (ground, excited & ionized states).

The following processes may result in the gain of excited atoms population, in any excited state (*L*).

a) Collisional excitation of neutral atoms by electrons whose energies re equal to or greater than the excitation energy of an atomic excited level (L). reaction in this process the electron energy is equal to $(\varepsilon_j + \varepsilon_{xL})$.
b) Excited atoms may be populated due to electron ion recombination process, reaction.
c) Also, their density may be increased due to dissociative recombination of dimer ions with electrons, reaction. In other words, excited atoms may be lost from any of the excited states.
d) Natural decay to the ground state with the emission of radiation. (Natural decay to any other levels is ignored in this model).

$$A^{*L} \rightarrow A + h\nu' \quad , \quad h\nu' = \varepsilon_{xL} \qquad (11)$$

e) Ionization by collision with electrons having energies greater than $(\varepsilon_i - \varepsilon_{xL})$, reaction.
f) Ionization by absorption of photons from the laser field, reaction the temporal variation of the ground state atoms due to the :
    a) Electron impact excitation and ionization.
    b) multiphoton ionization,
    c) Neutrals decay of excited atoms.
    d) formation of dimer ion through the reaction ,

$$2A + A^{+} \rightarrow 2A^{*} + A \,, \qquad (12)$$

Also their dissociative recombination, are all considered in the model.

The system of equations describing the various mechanisms leading to breakdown can be represented as follows.

The rate of change of the fraction of electrons f having energies between (ε) and $(\varepsilon + \Delta\varepsilon)$. During the time interval (Δt), due to the various possible interactions mentioned in the previous section is given by :

$$\partial f/\partial t=[\,G(\varepsilon)\,(\partial f/\partial \varepsilon)+2\varepsilon\,G(\varepsilon)\,(\partial^2 f/\partial \varepsilon^2)]+[B\{I(t)\}^k N(t)+\sum_L B_L\{I(t)\}^{kL}X_L(t)]+[\sum_L \nu_{xL}(\varepsilon+\varepsilon_{xL})\;N(t)\;f(\varepsilon+\varepsilon_{xL}+\nu_i(\varepsilon+\varepsilon_i)\;N(t)\;f(\varepsilon+\varepsilon_i)+\sum_L \nu_{ixL}\;(\varepsilon+\varepsilon_i-\varepsilon_{xL})X_L(t)\;f(\varepsilon+\varepsilon_i-\varepsilon_{xL})]-[\sum_L \nu_{xL}(\varepsilon)\;N(t)\,f(\varepsilon)+\sum_L \nu_{ix}(\varepsilon)\;X_L(t)\,f(\varepsilon)+\nu_t(\varepsilon)\,N(t)f(\varepsilon)]+D(\varepsilon)\;\nabla^2 f(\varepsilon)-R(\varepsilon)\,n_+(t)\,f(\varepsilon)\,f(\varepsilon)-r(\varepsilon)\;n_d(t)\;f(\varepsilon) \qquad (13)$$

The first bracket on the right hand side of this equation represents the rate of electron energy gain from the laser field, and the parameters included are as described by equations (1) and (2).

The second bracket represents the rate of electrons generation due to ionization of atoms (from the ground state or any excited state (from the ground state or any excited state L) by multiphotons absorption processes. $N(t)$ and $X_L(t)$ are the instantaneous density of atoms in their ground state and in any excited state (L).

$I(t)$, the instantaneous laser intensity, is given by equations [3]

$$I_{r,z}(t)=I_0(t)\;EXP\;\{-2\,(r^2/d^2+z^2/L^2)\} \quad \text{and}$$

$$I(t)=\hat{I}\;(t/\tau) \qquad \text{for } 0<t<\tau$$

$$=\hat{I}\;(2-t/\tau) \quad \text{for } \tau<t<2\tau$$

Where r and z are distances measured radically and axially from the focal spot, d is the spot diameter at the focal plane and L represents the distance between those points on either sides of the focal plane at which the intensity is half its maximum value Io at the focal plane, τ flash duration, $\hat{I}$ laser pulse of a trianglar shape with peak intensity.

And its exponent powers k and k_L are as defined in reactions (6) and (8), respectively.

The third bracket is the rate of appearance of electrons in energy range (ε) to ($\varepsilon + \Delta\varepsilon$) by the inelastic collisions (excitation and ionization collisions) with ground state atoms and ionization collisions with excited state atoms.

$\nu_{xL}(\varepsilon)$, $\nu_\iota(\varepsilon)$ and $\nu_{ixL}(\varepsilon)$ are the collisional rate coefficients of electrons , in any energy range ($\Delta\varepsilon$), of excitation , ionization of neutral and excited atoms, respectively.

The fourth bracket gives the loss rate of electrons, in the energy range (ε)to ($\varepsilon+\Delta\varepsilon$), as a result of excitation and ionization collisions with ground state atoms as well as the ionization of the excited atoms.

The last three terms are the loss rate of electrons in the energy range (ε) to ($\varepsilon+\Delta\varepsilon$) due to diffusion, recombination n a three body reaction, and dissociative recombination process respectively.

(D), (R) and (r) are the coefficients of electron diffusion three body recombination and dissociative recombination. n_+ (t) and n_d (t) are the density of ions dimer ions at any time t.

The growth rate of excited atoms (x_L), in any excited states L (L=1,2,..... z) , is given by :

$$\partial X_L/\partial t=\nu_{xL}\,N(t)\,n-B_L\,\{I(t)\}^k\,X_L-s_L\,X_L-\nu_{ixL}nX_L+Rn_+n^2+r\,n_d\,n \qquad (14)$$

The right hand of this equation is described as follows: The first term is the generation rate of these excited atoms due to excitation collisions between electrons, at different energy ranges. And ground state atoms. The second and third terms represent the loss rate of the

excited atoms from the excited state (L), through multiphoton ionization processes as well as due to their natural decays to the ground state. $s_L$in the third term is the transition probability of the excited state (L).

The fourth term is the loss rate of the excited atoms from the excited state L in collisional ionization processes with electrons in different energy ranges.

The last two terms are the generation rates of excited atoms due to electron recombination's collisions (three - body and dissociative).

Method of Calculation:

Equation (13) is a nonlinear equation which may be solved numerically in case of constant rate coefficients. Different methods can be used such as Euler, Adams prediction Corrector or the fourth - order Runge Kutta method. In using Euler method the density of electrons yielded at the end of the laser flash was too small. Runge - Kutta and Adams methods are extremely time consuming, thus they are not practicable.

Therefore equation (13) is evaluated numerically using a step- by - step integration method during short interval of time. The first and second derivatives in the elastic collision term (first bracket) are evaluated using a finite difference method such as:

$$\partial f/\partial t = [\, f_{j+1}(\varepsilon,t) - f_j(\varepsilon,t)\,] / \Delta\varepsilon \quad ,$$

and

$$\partial^2 f / \partial \varepsilon^2 = [\, f_{j+1}(\varepsilon,t) - 2\, f_j(\varepsilon,t) + f_{j-1}(\varepsilon,t)\,] / (\Delta\varepsilon)^2$$

$$\text{For } j=1,2,3,4,..... \qquad (15)$$

In using this finite difference method, it was found that the maximum value of Δt, which may be used in the integration, can be determined from the stability condition:

$$\Delta t \leq (\Delta\varepsilon)^2 / 2\varepsilon\, G(\varepsilon) \qquad (16)$$

It is obvious, from this equation, that (Δt) is not independent of time, i.e. it varies during the laser flash. This is shown from equation (2), where the laser intensity is assumed to have a triangular pulse shape. In this case, the obtained (Δt) is found to be so small compared with the laser pulse length.

Therefore, a large number of time steps ($\cong 10^{15}$ s) have to be computed in a single simulation of a breakdown experiment .

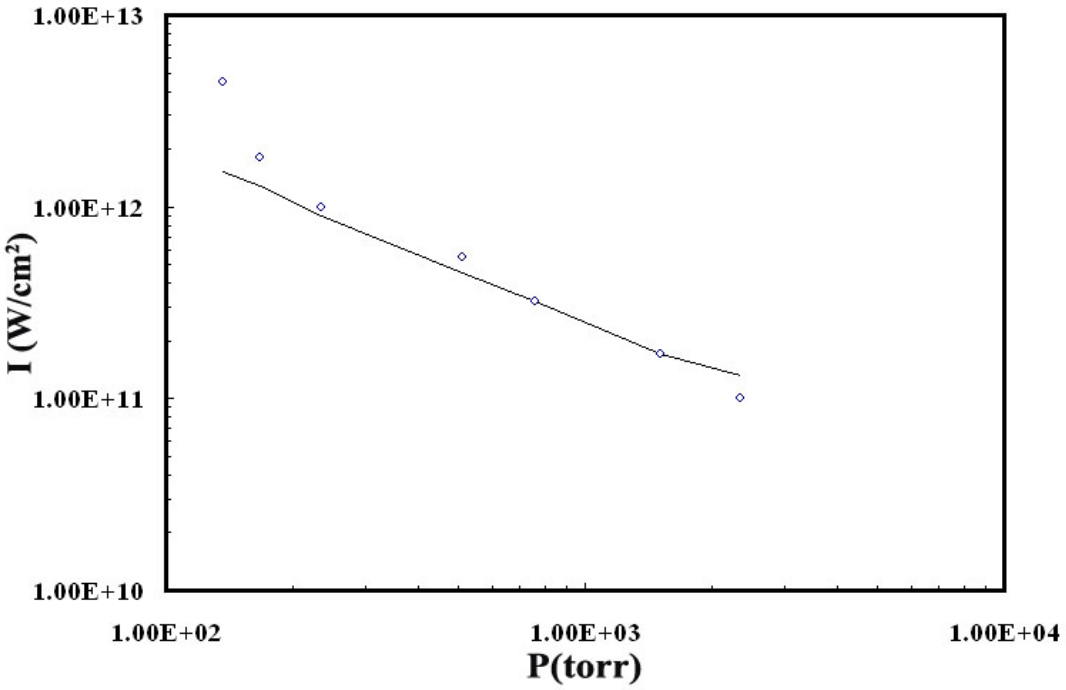
On applying this time criterion on the inelastic terms, the execution time for the program was found to be very much longer than it needs to be . In order to speed up the calculations, another time step (Δt_L) was also used. The values of these two time steps were adjusted slightly so as to make ($\Delta t_L / \Delta t$) an exact integer, (m). The effect of the elastic collisions was first calculated over a time – step (Δt), and this was repeated m times. Then the effect of inelastic collisions was calculated over the total time – step (Δt_L) .Since this calculation was done comparatively infrequently, a more elaborated formula could be used for the number of electrons involved in the ionization, excitation, and loss processes.

At any time t , number of electrons existing in the energy range ε to $\varepsilon+\Delta\varepsilon$ is f (ε) to f ($\varepsilon+\Delta\varepsilon$) . During an interval of time Δt_L this electron number will be changed as a result of the various inelastic collision processes considered in the model.

Similar expressions are applied to the other inelastic collision processes of ionizing the excited atoms. and exciting the neutral atoms, with the change of the threshold energy needed for each process. In case of the excitation process, n_o electrons are added to the lowest energy range, but the excited atoms are generated instead. Also the same ways of exponential decay are considered for the electron loss processes.

Results and discussion:

These are presented in figure [2] (solid line).



Figure[2] Compares between experimentally measured (circules) and calculated measured with present of all loses (solid line) .

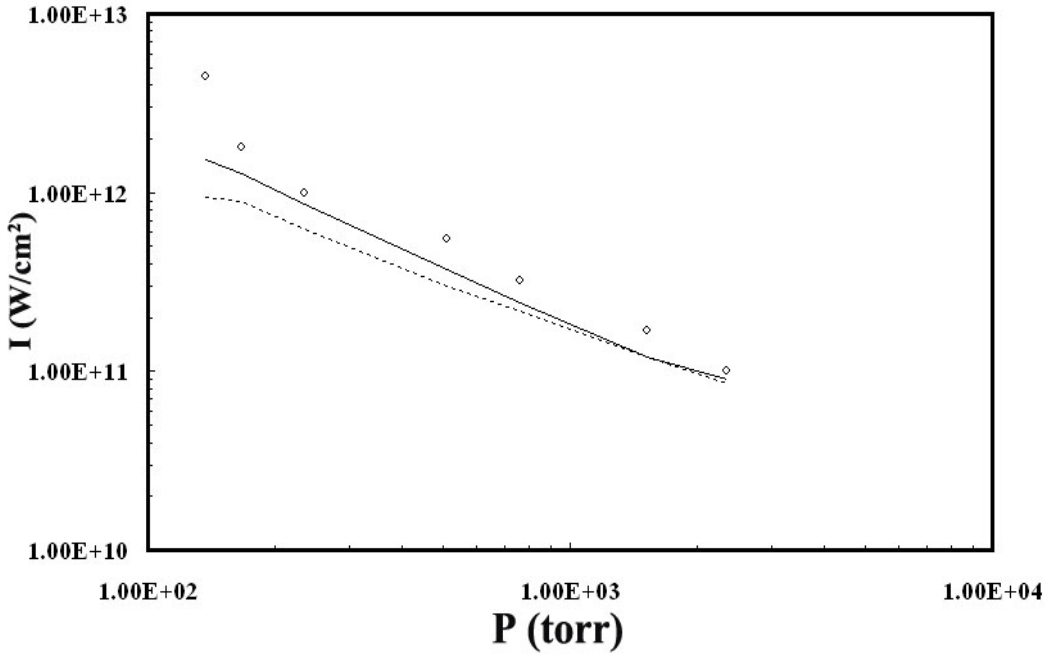
For an easy comparison the experimental thresholds of Rosen and Weyl [5], are also shown on the same figure (circles). It can be seen from this figure that a reasonable agreement is obtained between the calculated thresholds and the experimentally measured ones over the whole rang of gas pressure examined experimentally. In this figure no evidence of multiphoton ionization has been observed , but rather the results indicate that ionization proceeds via electron cascade mechanism , this provided by the strong dependence of threshold intensity on the gas pressure .

In calculating these threshold intensities, we have taken into account the effect of electron loss processes namely: electron diffusion and three body recombination in addition to the desiccative recombination. From our calculations we found that the desiccative recombination has almost negligible contribution to the breakdown phenomenon under the experimental condition considered in this analysis, therefore we remained only with the two electron loss processes.

In order to have an idea about the order of magnitude of these losses and their contribution to the breakdown phenomenon, calculations are carried out considering the separate effect of each of these loss processes.

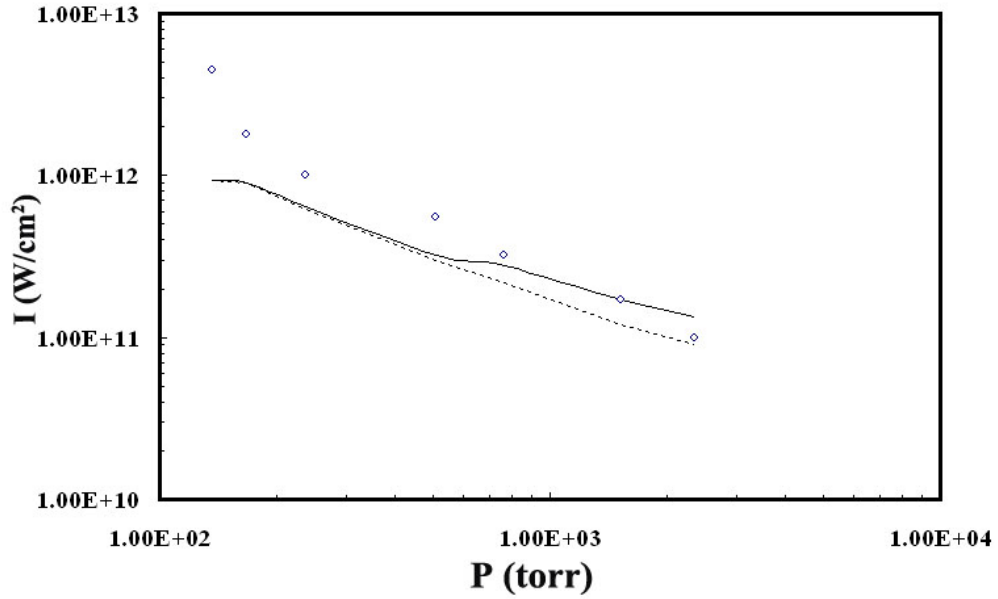
To examine the dependence of the loss process on the gas pressure, calculations are carried out for the threshold intensities as a function of pressure for two cases: i) When

electron diffusion is the only loss process . And ii) When three bodies recombination alone are considered.



Figure[3] comparison between experimentally result (circles) and calculated measured with present of diffusion loss (solid line) (no loss dashed line).

Figure (3) illustrates the calculated threshold intensity as a function of gas pressure when diffusion losses are present (solid line) , and when no losses are considered (dashed line) . The experimental thresholds are also shown in this figure (circles) to illustrate the effect of diffusion losses on the breakdown phenomenon. From this figure it is clear that diffusion losses are acting very effectively at low pressures, while it has an almost a negligible contribution at pressures above the atmospheric. This is observed from the coincidence of the solid line (diffusion) and the dashed line (no loss) at high pressures, where these two curves show a considerable deviation from the experimentally measured thresholds.



Figure[4] comparison between experimentally result (circles) and calculated measured with present of recombination loss (solid line) (no loss dashed line) .

Similarly in figure (4) the solid line represents the calculated thresholds when three bodies recombination losses are the only electron loss process considered ,while the dotted line represents the case when no losses are present . It's also clear from this figure that recombination losses are more effective at high pressures. The coincidence of the two curves bellows the atmospheric pressures indicates the negligible contribution of this loss process at these pressure regimes.

To give deeper insight into the effect of loss process on the breakdown phenomenon the electron energy distribution function and its parameter are calculated at two different values of gas pressure ($2.356x10^2$ and $2.35x10^3$ torr) , for the separate contribution of the two loss processes considered .These will be refereed in the following as case(1) ($P=2.356x10^2$ torr) and case (2)($P=2.35x10^3$ torr) respectively, these two values have been chosen deliberately since they lie in two different pressure regions. Therefore calculations are carried out, for each case to study the separate contribution of each loss process.

Table (1) Table of values that have been used to draw graphs.

I (W/cm2)	P (torr) Experimentally result	P (torr) Calculated measured with no loss	P (torr) calculated measured of diffusion loss	P (torr) calculated measured of recombination loss	P (torr) calculated measured with present of all loses
$1.37*10^2$	$4.50*10^{12}$	$9.40*10^{11}$	$1.53*10^{12}$	$9.40*10^{11}$	$1.53*10^{12}$
$1.67*10^2$	$1.80*10^{12}$	$9.00*10^{11}$	$1.28*10^{12}$	$9.00*10^{11}$	$1.28*10^{12}$
$2.36*10^2$	$1.00*10^{12}$	$6.20*10^{11}$	$8.60*10^{11}$	$6.37*10^{11}$	$9.00*10^{11}$
$5.09*10^2$	$5.50*10^{11}$	$3.00*10^{11}$	$3.70*10^{11}$	$3.20*10^{11}$	$4.50*10^{11}$
$7.60*10^2$	$3.20*10^{11}$	$2.16*10^{11}$	$2.40*10^{11}$	$2.80*10^{11}$	$3.20*10^{11}$
$1.52*10^2$	$1.70*10^{11}$	$1.20*10^{11}$	$1.20*10^{11}$	$1.70*10^{11}$	$1.70*10^{11}$
$2.35*10^2$	$1.00*10^{11}$	$8.60*10^{10}$	$9.00*10^{10}$	$1.33*10^{11}$	$1.33*10^{11}$

Conclusions

The following conclusion could be obtained:

- The dissociative recombination has almost a negligible contribution to the breakdown phenomenon under the experimental conditions considered in this analysis .
- The diffusion losses are very effective at pressure values below atmospheric , during the first stage of the laser flash .
- Three body recombination's are acting at high pressure (above atmospheric) during the second half of the laser flash .
- We also conclude that the new inelastic processes such as photionization and electron impact ionization of the electronic excited states (4s and 4p) play an important role to the phenomenon associated with laser induced breakdown of argon gas .

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