Studying the Role of the Physical Processes dependence on gas pressure in The Breakdown of Molecular Oxygen by 1064 nm Laser Radiation

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Abstract: This work presents an investigation of the experimental measurements that carried out to study the breakdown threshold dependence on gas pressure in the breakdown of molecular oxygen by a Nd: YAG laser source operating at wavelengths 1064 nm with a pulse duration of 5.5 ns over a gas pressure range varies between 190-3000 torr. The study is devoted to find out the role played by the physical processes in determining the threshold intensity of molecular oxygen breakdown as a function of gas pressure. In doing so a previously developed electron cascade model is modified and applied. The modification assigned the inclusion of an electron diffusion term to account for electron losses out of the focal volume on the experimentally tested low pressure regime. Besides, the model takes into account most of the physical processes which might take place during the interaction between the laser beam and the oxygen molecules. The result of computations showed reasonable agreement between the calculated threshold intensities and the experimentally measured ones over the tested gas pressure range. This proves the validity of the model. In addition the study of the performed of the electron energy distribution function, EEDF, and its correlated parameters viz, temporal evolution of: electron density, excitation rate, ionization rate and electron mean energy as well as the variation of the EEDF during the laser pulse verified the exact correlation between gas pressure and the physical process responsible for the production and loss of electrons or their energy during the breakdown phase.

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1. Introduction

Oxygen is one of the most important constituent of atmospheric air as well as it gives an example of the greatest effective electronegative gas which plays an important role in the earth's atmosphere. Although numerous studies on the breakdown of molecular oxygen up to the atmospheric pressure have been performed by many authors (see for examples Kroll and Watson, 1972, Dewhurst, 1978, Sticker and Parcker, 1982, Gamal *et al.,* 1987, Tambay and Thareja, 1991 Davis *et al.,* 1991 and Sircar *et al.,* 1997) there is a still inadequate explanation of this phenomenon at both very low and high pressure values.

Therefore, this work presents a numerical investigation for the breakdown threshold behavior as a function of gas pressure exposed experimentally in the breakdown of molecular oxygen by Nd: YAG laser source operating at the wavelength 1064 nm with a pulse duration of 5.5 ns over a gas pressure range varies between 190-3000 torr. These experimental conditions correspond to the measurements given by Phuoc (2000).The study intended to determine the correlation between threshold intensity and electron production and loss processes as a function of gas pressure.

In doing so a previously developed numerical model Evans and Gamal (1980) is modified and applied. The modification was mainly concerned with the inclusion of an electron diffusion term to account for electron losses at the experimentally tested low pressure values.Besides, the model take into account most of the physical processes which might take place during the interaction between the laser beam and oxygen molecules (Gamal and Omar, 2001). The model considers the inverse Bremsstrahlung absorption as a process that leads to the gain of energy of free electrons from the electric field associated with the laser beam. In addition, it takes also into account electron collisional processes to excite neutral molecules as well as to ionize ground and excited molecules. The computations are performed to find out relations between threshold intensity and gas pressure, the Electron Energy Distribution Function (EEDF) and its parameters to determine the exact contribution of each physical process to the breakdown phenomenon at each gas pressure value.

2. Theoretical Formulation

2.1 Basic equation

A detailed description of the electron cascade model is given in Evans and Gamal (1980). The model is modified by Gamal and Omar (2001) to suit the case of molecular oxygen. In this modified model the oxygen is treated as a five level molecule ground state, three electronic excited states and an ionized state (Kroll and Watson, 1972). Accordingly the following physical processes are taking into account: (i) Electron inverse-Bremsstrahlung absorption.(ii) vibrational excitation losses,(iii) Electron impact excitations iv) Electron impact ionization of ground state molecules by electrons,(v) Collisional ionization of the three excited states by electrons (vi) Photo-ionization of the excited states, (vii) Two- and three-body attachments.

Accordingly, the modified time dependent Boltzmann equation solved for the electron energy distribution function f (ε,t) is written as:

$$\frac{\partial f(\varepsilon,t)}{\partial t}=\frac{2\varepsilon}{3m\nu_m(\varepsilon)}\nabla^2 f+\frac{1}{3}\varepsilon_0\nu_m(\varepsilon)\frac{\partial f}{\partial \varepsilon}+\frac{2}{3}\varepsilon_0\nu_m(\varepsilon)\frac{\partial^2 f}{\partial \varepsilon^2}+\frac{\partial(Q_v f)}{\partial \varepsilon}+\textit{Inelastic collision terms} \quad (1)$$

Where f(ε,t)dε represents the number density of electrons with energy between ε and ε+dε, $\varepsilon_o = e^2 E^2 / 2m\omega^2$ is the average oscillatory energy of an electron in the laser field with electric field amplitude E and angular frequency ω, $\nu_m(\varepsilon)$ is the momentum transfer collision frequency and Q_v is the rate of transfer of energy from an electron of energy ε to vibrational levels.

In this equation, the first term on the right-hand side represents the rate of electrons loss from the radiated volume due to diffusion. Here, we shall follow the custom to rewriting this by setting $\nabla^2 f = -\Lambda^{-2} f$, where Λ is the characteristic diffusion length. This length characterizes the distance over which a particle should diffuse in order to be lost from the plasma. Following standard optical focal theory, the minimum focal volume is assumed to be cylindrical (MacDonald 1966) of radius $r_0 = f_l\ (\alpha /2)$ and axial length $l_0 = (2\ \sqrt{2}\ -1)(\alpha/d) f_l\ 2$, where f_l is the focal length of the lens, d is the diameter of the unfocused laser beam and α the corresponding beam divergence(Kroll and Watson, 1972). The second term expresses the electron energy gain; the third term is referred to diffusion of electrons along the energy axis, while the third term describes the elastic energy loss by colliding electrons through vibrational excitation. The inelastic collision terms include electronic excitation and ionization of ground state molecules, in addition to the photo-ionization and electron impact ionization of the formed excited molecules. It also comprises the electron loss processes through electron attachments.

2.2 Data of oxygen molecule

In this model for oxygen molecule we used the same empirical formulae for rate constants and coefficient given by Kroll and Watson (1972) and applied in our previous paper Gamal and Omara (2001). These represent: the collision frequency, excitation of vibrational levels, collisional excitation of the electronic levels, collisional ionization, stepwise ionization which includes electron impact ionization and photoionization of the formed excited molecules. In this analysis, the photoionization coefficients for the three excited states of the oxygen molecule are calculated applying the formula given by Grey Morgan (1975) and shown in table (1). The rate coefficients of the two and three-body attachment are also taking from our previous paper (Gamal and Omara, 2001). Since the investigated measurements are performed at low pressure regime so it was necessary to include into the model the loss of electrons out of the focal volume due to diffusion process. The rate of this process is taken as

In this work the rate of diffusion losses is defined as:

$$A_D(\varepsilon)=\left(\frac{2\varepsilon}{3m\nu_m}\right)\Lambda^{-2} \quad \mathbf{(2)}$$

Here, the characteristic diffusion length Λ is considered to be energy independent. Adopting the experimental conditions considered in this analysis (Phuoc, 2000), where the beam mode of the laser pulse is taken to be close to Gaussian, so that the near focus may approximately be expressed in a cylindrical shape of radius r0 and length l0. Thus the characteristic diffusion length is written as

$$\Lambda^{-2}=\left(\pi/l_0\right)^2+\left(2.40/r_0\right)^2 \quad (3)$$

2.3 Method of Calculation

Numerical computations have been carried out by solving equation (1) using a step-by-step integration. Since the ionization energy of oxygen is 12.06 eV, the step-length in energy,ΔE, was chosen so that the complete energy distribution could be represented by about 25 equally-spaced points. The derivatives $\partial f/\partial t$ and $\partial^2 f/\partial t^2$ were evaluated by the finite difference technique. The inelastic terms could have been included as difference terms in the basic differential equation, but there are good reasons for treating them separately (Evans and Gamal, 1980). The breakdown criteria used in this analysis is taken as the attainment of a degree of fractional ionization $\delta \approx 0.1$ % of the neutral gas molecules present in the focal volume.

The instantaneous temporal variation of the laser intensity I (t) at any time, t, is assumed to have a Gaussian shape, and written as

$$I(t) = I(o)A(e^{\frac{-(t-\tau)^2}{\tau^2}} - B) \quad (4)$$

Where, τ is duration pulse, A, B are constants.

To avoid the discontinuity of the EEDF, the calculations started with one electron having a Gaussian energy distribution with a mean energy of 4 eV. In addition, the depletion of the ground state as well as the excited states due to ionization and excitation is also taken into account.

3. Results and discussion

Calculations are basically devoted to investigate the comparative contribution of the physical processes to the breakdown of molecular oxygen over pressure range extended from 190 to 3000 torr irradiated by 1064 nm laser radiation with pulse duration 5.5 ns. In doing so, computations are started by determining the threshold intensity as a function of gas pressure and plotted in figure (1) (solid line). For an easy comparison the experimentally measured thresholds (Phuoc, 2000) are also shown on this figure (Solid squares).This comparison showed a reasonable agreement between the calculated thresholds and the measured ones which validates the model to elucidate the breakdown of oxygen under the experimental conditions applied in this analysis. It is also noticed here that both calculated and measured threshold intensities showed different behaviors over the tested pressure range, where at the low pressure region it suffers a noticeable decrease up to 400 torr. Thereafter its values showed very slight changes up to 1500 torr followed by a drop over the higher pressure regime (2250-3000 torr).This behavior indicates that breakdown occurs through different mechanisms over the different pressure regimes. Therefore, it was necessary to perform a comprehensive study on the exact correlation between gas pressure and the physical processes responsible for oxygen breakdown. In doing so, calculations are carried out to obtain the EEDF and its parameters at the experimentally examined gas pressures. Curves 1 to 6 on figure (2) correspond to the EEDF calculated at the peak (a) and end (b) of the laser pulse over pressure range from 190 to 3000 torr respectively. It is noticed from figure 2a that the EEDF varies unsystematically over the whole pressure range. Since its value increases with the gas pressure over the low pressure range as shown by curves 1and 2, while it decreases with the increase of the gas pressure up to 2250 torr (curves 3-5).At the higher pressure a sudden increase of the EEDF is noticed with its tail attains higher values up to the energies higher than the ionization thresholds, as shown by curve (6). This behavior indicates that at the lower pressure electrons may be lost through electron diffusion process. At the high pressure however electrons may lose their energy due to the inelastic collisional processes leading to an increase of the low energy electrons (notice the high values of the EEDF at zero electron energy curves 3-6). The behavior shown by curve (6) assures the high rate of collisional ionization which can easily overcomes electron losses due to attachment processes. The fast decrease of the EEDF shown at the low energy region reveals the important role played by vibrational losses in depleting the low energy electrons.

At the end of the laser pulse (Figure 2b) the EEDF is found to decrease with the increase of the gas pressure as shown by curves (1-6).This decrease is more pronounced at the higher pressures (Curves 3-6).This behavior corresponds to a strict loss of energy and to a variation of the number of free electrons, due to the globally elastic and inelastic collisional processes encountered in this model such a vibrational and electronic excitations and attachment processes.

The results of this study showed that at the low pressure regime ionization are mainly proceeds via multi photon ionization processes. At the higher pressures, however, collision ionization processes are the main source of electron generation.

The study of the time evolution of represented in figure (3) showed high growth rate of the electron density at the low pressures. This is an evidence for the effective contribution of the multi photon ionization process. On the contrary the slow growth rate of the electrons during the early stages of the laser pulse for the high pressure region confirm the importance of the electron collision process in the breakdown of gas.

For deeper understanding of the role played by the physical processes involved in the model to the breakdown of oxygen, a study is carried out to obtain the time evolution of the electron density as a function of gas pressure. This relation is illustrated by the curves (1-6) in figure (3).The rapid growth of the electron density at the low pressure region (Curves 1-2) assures that at these pressures electrons are mainly generated through photo ionization processes. On the other hand curves (3-6) showed a very slow growth of the electron density during the early stages of the laser pulse. This in turn gives an evidence for the importance of collisional ionization in enhancing the electron growth rate which is confirmed by the exponential increase of the electron density shown over the time interval 1.0-3.0 ns. Moreover the decrease of the electron density at the end of the laser pulse shown by curves (4-6) reflects the effect of electron losses through attachment processes.

Since excited molecules are found to play important role in gas ionization, at the low pressure values (through photoionization process), and high pressures (through collisional ionization process), it was necessary to study the correlation between temporal variation of excitation and ionization rates at the different values of gas pressure. These are represented by curves (1-6) in figures (4, 5). From these figures it is noticed that the results obtained at the low pressures (Curves 1,2) show a contradictory behavior, since over the early stages of the laser pulse these curves, showed decrease in figure (4) and an increase in figure (5).This behavior illustrates that, the depletion of excited molecules results in fast growth of ionization rate through photoionization process. The effect of this process is contested with electron collisional ionization of ground and excited molecules as the gas pressure increases. This is clear from curves (3 and 4) on figure (4) where the excitation rate showed a deep well at 1.0 ns followed by a sharp increase to a relatively high value which is continued up to the end of the laser pulse. This result showed opposing behavior in figure (5) for the same curves where a significant peak appeared at 1.0 ns followed by a decrease over the second half of the pulse. On the higher pressure region however, electron density growth is very pronounced on the time interval 1.0-2.0 ns thereafter its value shows a noticeable decrease (curve 5). This behavior is clearer at pressure 3000 torr as shown by curve (6). The fast decrease of the ionization rate shown by curves (5and 6) on figure (5) confirms the high rate of electron losses due to attachment processes during the second half of the laser pulse.

To specify precisely the correlation between gas pressure and the ionization process responsible for electron density growth figure (6) represents a relation for the time variation of the electron mean energy at the different values of the gas pressure as shown by curves (1-6). From this figure it is clear that the electron mean energy behaves differently on the low pressure regime where curves (1 and 2) lie considerably above those curves which represent the higher pressure region (curves 3-6).This behavior proves that ionization on this regime takes place via photoionization mechanism which is pressure independent. The drop shown by curves (1 and 2) during the early stage of the laser pulse indicates the effect of electron diffusion losses. Meanwhile the constant value observed on the remaining time of the pulse gives an evidence for the slow ionization rate shown by the same curves in figure (5).On the contrary, at the high pressure region where collisional ionization starts to have a pronounced contribution to the electron density growth, the obtained temporal variation of the electron mean energy followed almost the variation of the laser intensity. This behavior assures the high probability of collisional processes over the pressure region which is extended from 760 torr to 3000 torr (Curves 3-6). Moreover, it clarifies also that breakdown and plasma generation starts only near the end of the laser pulse where enough electron density can be present at this time interval.

For completeness a study is performed for the time variation of the EEDF at the tested pressure values. This study proves that over the tested pressure range the EEFD has a non Maxellian form where its shape varies during the laser pulse. In addition its variation depends mainly on the gas pressure,since at the low pressure region (Figure 7 i and ii) its value increases rapidly during the early stages of the pulse up to its peak where a slow growth is observed leaving high values of the EEFD up to its tail. This means that high electron density can attain high energy during the laser pulse. This is confirmed by the contour representation of the EEDF which gives a deeper appearance on the plasma parameters such as: formation time, electron density and amount of energy carried by the generated electrons. At low pressures figure (7i and ii) creation of breakdown is observed at the end of the pulse. This behavior is opposed as the gas pressure increases where the breakdown is initiated before the end of the pulse and moves towards the pulse peak as the gas pressure increases. This action assures the significant contribution of collisional ionization processes. It is also noticed her that in the breakdown region most of electrons attain only low amount of energy since they lose their energy in inelastic collisions as shown in figure (7iii and vi). To clarify this results this relation is plotted with small electron energy scale and represented on the same figure. Notice the confined breakdown region represented by the red color surrounded by ionization zones signified by a color bar extended from orange to black that correspond to the degradation of the electron density through the successive zones. Moreover this figure shows the effect of electron losses by diffusion at the lowest pressure value and the high rate of attachment losses at the highest one.

Table 1 Calculated values of the photoionization rate coefficients and related parameters

Wavelength (nm)	Photon energy (eV)	Excited level energy (eV)	K (number of Photons)	Absorbed energy (eV)	Ionization energy (eV)	Residual energy (eV)	Photo-ionization Coefficient ($cm^{-2}W^{-1})^{K}S^{-1}$)
1064	1.17	4.5	7	8.16	7.58	0.60	3.5×10^{-93}
		8.0	4	4.66	4.06	0.60	6.1×10^{-46}
		9.7	2	2.36	2.34	0.03-	1.0×10^{-15}

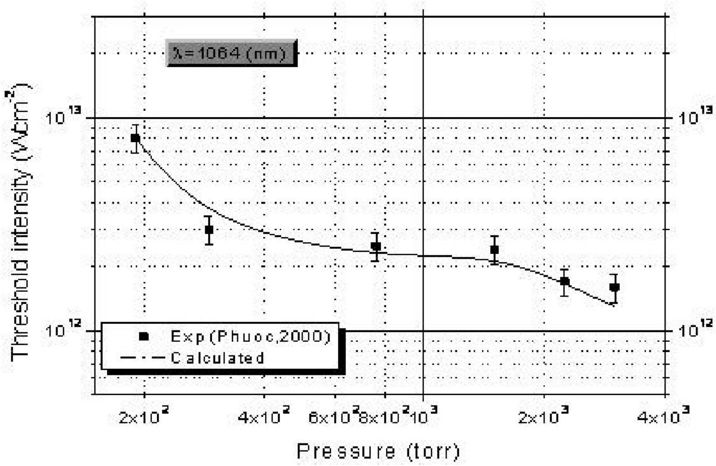

Fig.1 Comparison between the calculated and measured threshold intensity as a function of gas pressure

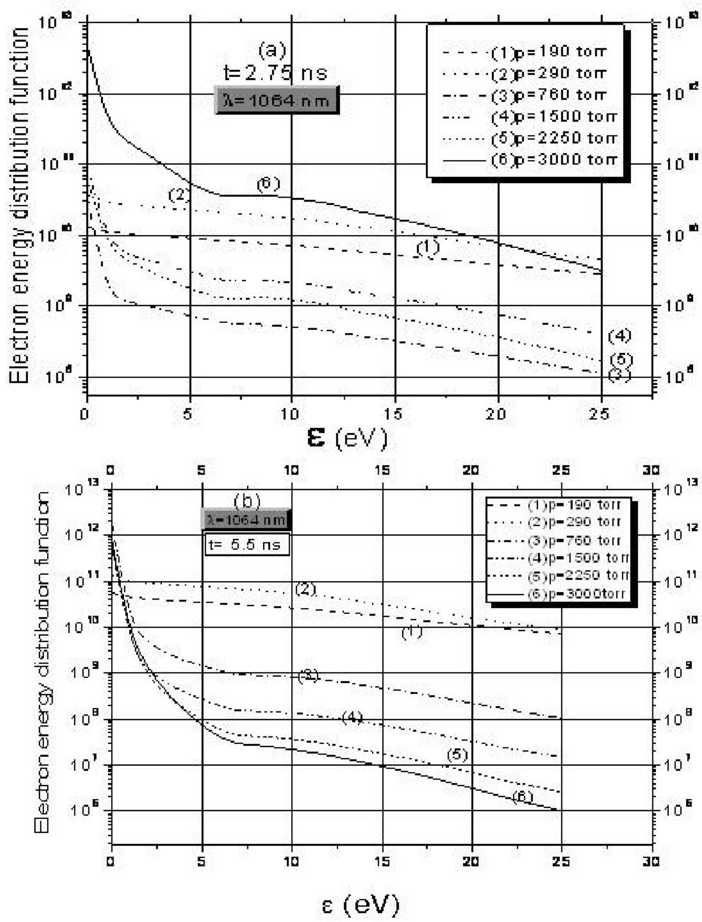

Fig.2 EEDF calculated at the (a) peak and (b) end of the laser pulse at different values of gas pressure

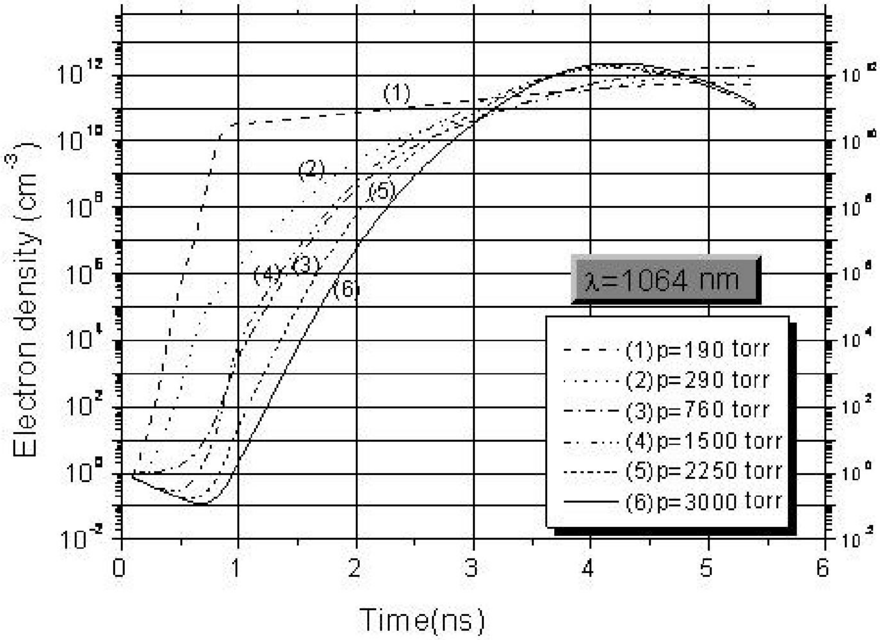

Fig.3 Time evolution of the electron density calculated for the different pressure values

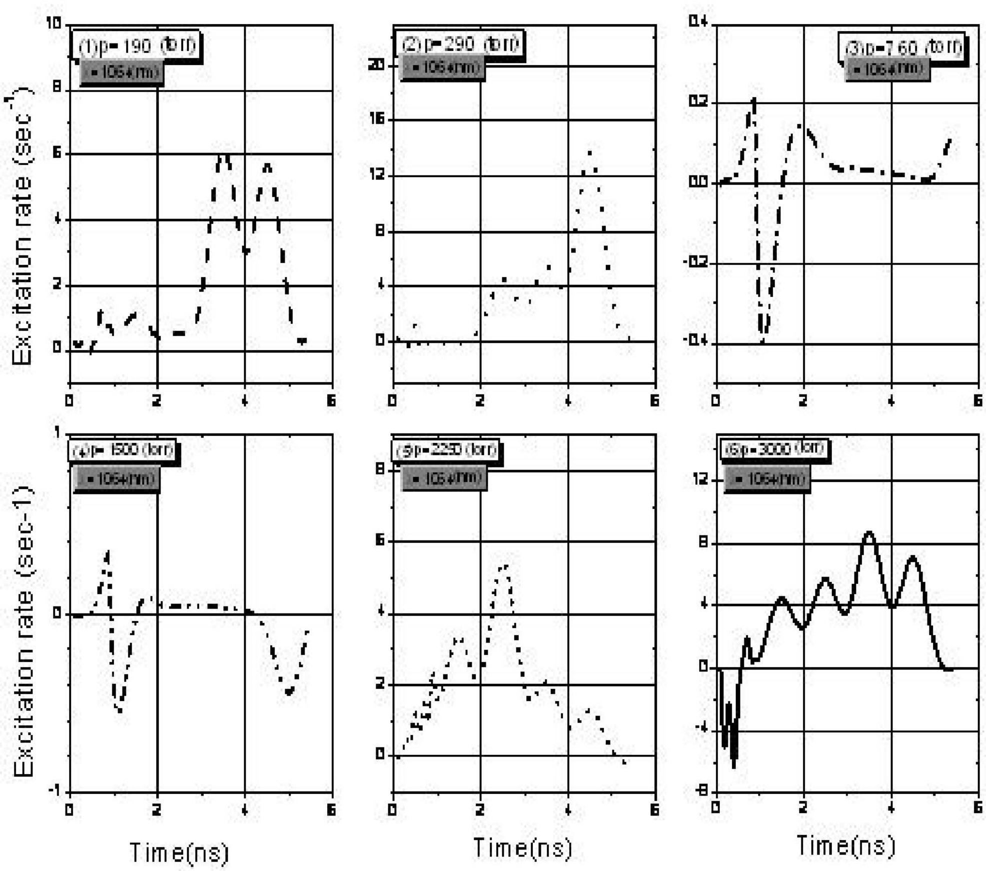

Fig.4 Time variation of the excitation rate obtained for the different values of the gas pressure

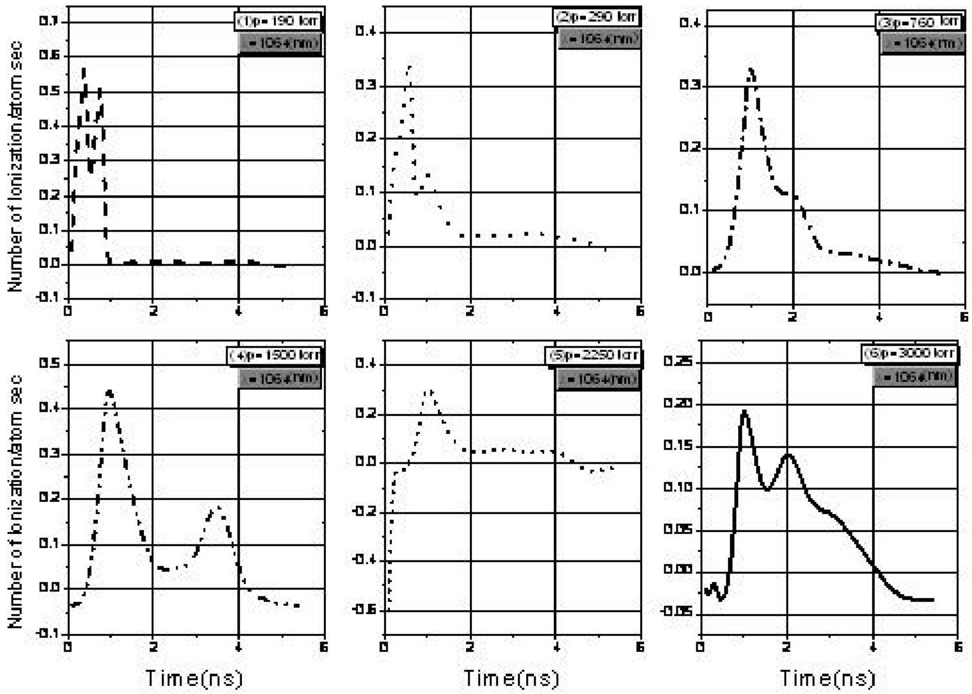

Fig.5 Time variation of the ionization rate calculated at different values of the gas pressure

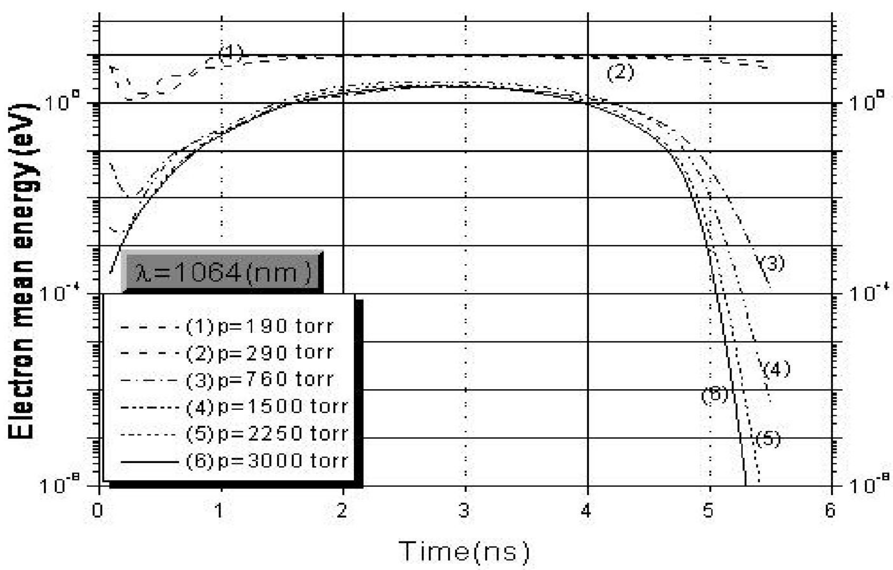

Fig.6 Electron mean energy plotted against time at different values of the gas pressure

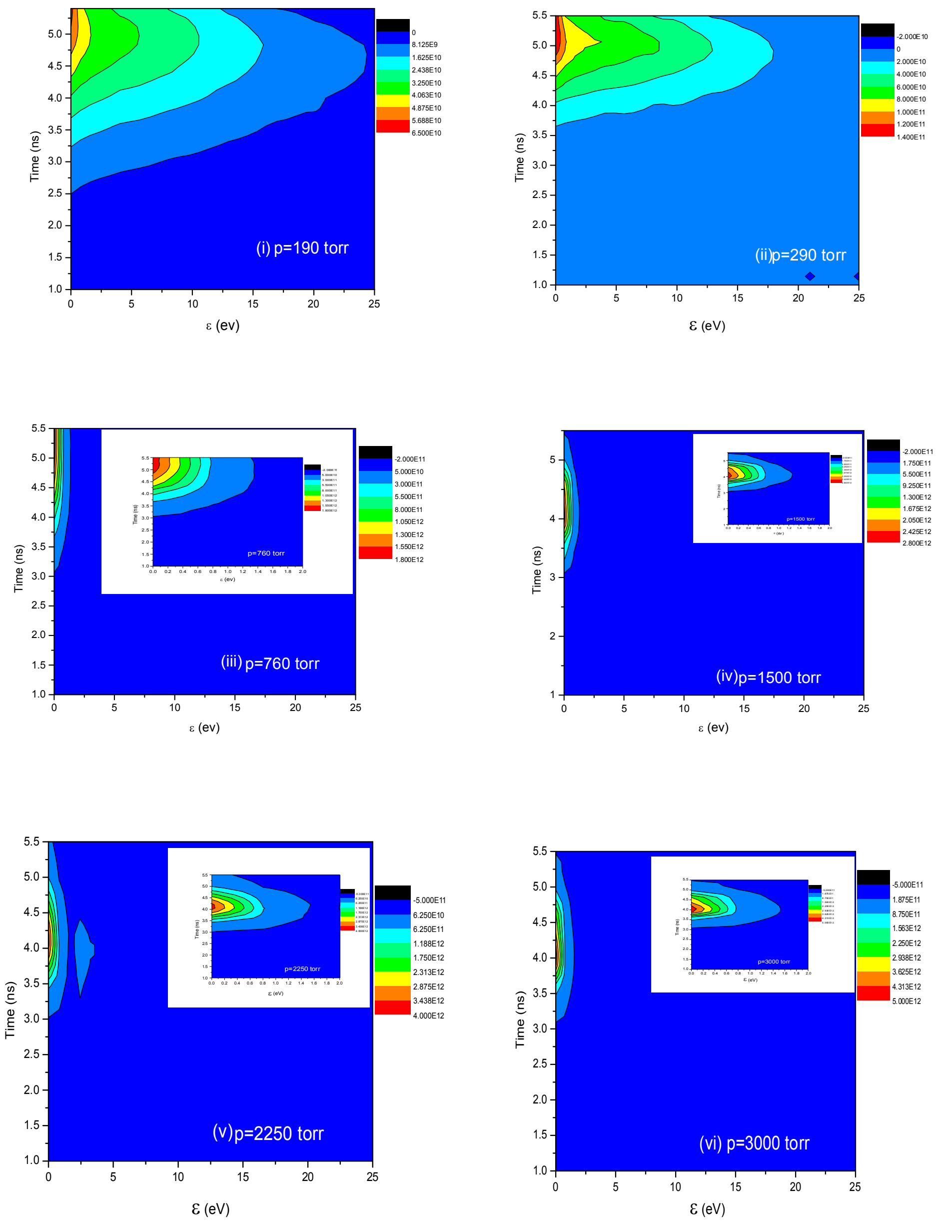

Fig.7 Contour representation of the time variation of the EEDF calculated for the different gas pressures.

4. Conclusion

In this work a numerical electron cascade model (Gamal and Omara, 2001) is modified and applied to investigate the correlation between gas pressure and physical processes (involved in the interaction) and their effect on the determination of the breakdown threshold intensity of molecular oxygen under the experimental conditions given by Phuoc (2000).In this measurements molecular oxygen over a pressure range 190 torr – 3000 torr is irradiated by laser radiation operating at wavelength 1064 nm and pulse duration of 5.5 ns. The results of this study proved that at the low pressure region (190 torr -290 torr) electrons are mainly generated through photo-ionization of the formed excited molecules. Diffusion losses act to deplete the electron density over this pressure range. On the intermediate pressure region (760 torr -1500 torr) ionization proceeds essentially via the combined effect of photo-ionization and collisional ionization of the formed excited molecules. Loss processes have only slight contribution over this pressure range. At the higher pressure range (2250 torr

-3000 torr) collisional ionization dominates with extremely high probability. Inelastic collisions due to electron impact excitation and ionization are responsible for the breakdown of molecular oxygen and hence the low values of the threshold intensity observed experimentally and verified numerically. Electron attachment losses (two and three body attachment) are operating with high rate over this pressure range.

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