Theoretical investigation of argon plasma formation induced by laser radiation

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Abstract: A previously developed electron cascade model is modified and applied to investigate the breakdown threshold for plasma formation and propagation in the focal volume . The study is devoted to investigate the measurement that carried out on the breakdown of argon over a pressure range 0.013-1.00 atm (10 -760 torr) induced by 532 nm of Nd:YAG laser with pulse length 8 ns and maximum energy 500 mJ. The model solves numerically the time dependent Boltzmann equation and set of rate equations that describe the change of the excited states population .The result showed good agreement between the calculated threshold intensities/ or laser input energy and the measured ones over the tested pressure range, this in turn validate the applied numerical model. More over the calculation of the EEDF and its parameters showed the correlation between gas pressure and physical processes responsible for the gas breakdown and plasma formation . Taken into consideration the spatial and temporal variation of the laser intensity in the focal volume it was possible also to present in this work the study of the effect of laser input energy on plasma propagation along the axial distance of the spatially varying focal volume. The result of this study illustrated the increase rate of plasma propagation by increasing the input energy, where it is found that at input energy equals three and half time its threshold energy value, the plasma propagates to cover the whole Rayleigh range in the backward direction.
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1. Introduction

The phenomenon of laser induced breakdown and plasma generation in gases have been studied extensively both experimentally and theoretically during the last five decades. Recently ,this phenomenon found a great importance for various applications, which include micro industries in electronics , environmental application for the measurement of pollution, surface cleaning, besides its application in medicine and biology. These studies showed that such applications are mainly depend on the characteristics of the plasma formed in the breakdown region. One of the main features of the formed plasma is its propagation in the backward direction of the focal volume as the input laser energy exceeds the threshold energy for breakdown. Moreover as the gas pressure increases the rate of propagation increases and more absorption of the input energy occurs in the plasma causing less transmission in the forward direction. Therefore more interest is devoted to study the physical processes responsible for this propagation (Yamada *et al.,* 1985;Yamada *et al.,* 1994;Tsuda *et al.,* 1997;Mlejnek *et al.,* 1999 ;Bindhu *et al.,* 2003). In these studies it was found that these physical processes depend on the parameters of the laser source as well as the nature of the irradiated gas.

Accordingly , A theoretical study of the phenomenon of laser induced breakdown and plasma generation in argon under the experimental conditions given by Bindhu *et al.* (2003) is presented . In this experiment the breakdown is obtained using a focused high intensity laser beam of wavelength 532 nm , pulse duration 8 ns and maximum energy 500 mJ , to irradiate argon gas over a pressure range varies between 10-760 torr, which is equivalent to 0.013-1.0 atm .This gas has been chosen since it has examined experimentally by various authors see for example: Yamada *et al.,* 1985;Yamada *et al.,* 1994; Tsuda *et al.,* 1996; Mlejnek *et al.,* 1999 ;Bindhu *et al.,* 2003). Moreover ,it showed a minimum Ramsuer, in the relation between the momentum transfer collision cross-section and the electron energy. This minimum might has a noticeable effect on rate of energy gain by electrons from the laser field during the Inverse Bremsstrahlung absorption process which plays an important role in the breakdown of argon (Gamal *et al.,* 1986). The investigation is based on a modified electron cascade model (Gamal and Azzouz 1987; Gamal *et al.,* 1999) which depends on the numerical solution of the time dependent Boltzmann equation simultaneously with a set of rate equations that describe the rate of change of the formed excited states density. Calculation assumed first temporal variation of the laser intensity in a cylindrical focal volume with a Gaussian shape .This enables the study of the threshold intensity as a function of gas pressure. In addition a study of the temporal variation of the electron energy distribution function (EEDF) and its parameters during the laser pulse as a function of the gas pressure demonstrated the correlation between the physical processes responsible for the gas breakdown. The study takes also into account the effect of electron gain and loss processes at the different gas pressure values. To study the plasma propagation in the focal volume the model considered the temporal and spatial variation of the laser intensity along the axial and radial axes of the focal volume as a function of the laser input energy. This study clarifies the relation between the density of the formed plasma and its absorption rate of the laser input energy as well as its propagation rate in the focal volume.

2. Theoretical formulation

2.1 The model

A detailed description of the model is given in Evans and Gamal (1980) ; Gamal and Azzouz (1987) and Gamal *et al.* (1999). Here we summarize only the outlines of the model. The model is based on the assumption of the presence of at least one free electron in the focal volume at the onset of the laser pulse. This electron gains energy due to collision with neutral gas atoms in the presence of laser radiation through inverse Bremsstrahlung absorption. The energy gained from the laser field by electrons is given by, inelastic collision terms (1)

$$\frac{\partial n(\varepsilon,t)}{\partial t} = \frac{1}{3}\,\varepsilon_o\,\nu_m(\varepsilon)\frac{\partial n}{\partial \varepsilon} + \frac{2}{3}\,\varepsilon_o\,\varepsilon\,\nu_m(\varepsilon)\,\frac{\partial^2 n}{\partial \varepsilon^2} +$$

where ε0 =e2E2 /2mω2 is the average oscillatory energy of an electron in the laser field with electric field E and angular frequency ω, e and m are the electronic charge and mass, νm(ε) is the momentum transfer collision frequency and n(ε) is the electronic density at energy range ε , ε+dε.

The first term in the right-hand side corresponds to energy gain, the second term results in no net energy gain and is referred to as the energy diffusion along the axis.

Adopting the same assumption for the argon atom model given by Weyl and Rosen (1985), therefore, the argon atom is treated as a four-level atom such as: a ground state; an excited state 4S state at an energy loss 11.6 eV; an excited state 4P at an energy loss of 13.2 eV; and an ionized state at energy 15.75 eV.

Accordingly, the following inelastic collisional and radiative processes are taken into account: (1) Inverse Bremsstrahlung absorption; (2) electron impact ionization of ground state atoms with electrons having energies >15:8 eV; (3) electron impact excitations to the 4S and 4P states with electrons having energies > 11:6 eV and 13.2 eV, respectively; (4) photo-ionization of these formed excited states; and (5) collisional ionization of the excited states by electrons having energy > 4:65 eV for the 4S state and 2.85 eV for the 4 P state, respectively. Beside these excitation and ionization processes, loss processes such as electron diffusion out of the focal region and electron-ion recombination processes (three body recombination) are also included in this analysis.

2.2. Argon Cross sections and rate coefficients

The various relevant cross sections and rate coefficients of the argon gas considered in the present work were as follows.

The collision cross section of the momentum transfer for argon is taken from Gamal *et al.* (1986) where a curve fit expression was obtained using the experimental data giving by Hayatshi (1981) as

$_m=10^{-15}/(1+144.6356\quad -2583.08\quad ^2 + 35035\quad ^3$ $-85386.8\quad ^4)$ for 0.0< < 0.3 eV (2)

and

$_m=10^{-17}[11.467(\quad e^{-0.093}) + 2{:}817(\quad e^{-0.093})^2$ $+0.693(e^{-0.093})^3]$ cm^2

-Then the collision frequency n_m is related to the collision cross section s_m by the following relation

$_m = N \quad _m(2 \quad /m)^{1/2}$ s^{-1} (4)

where N is the gas density per unit volume.

-For cross sections of excitation and ionization, we applied those which were considered by Weyl and Rosen (1985) as:

$_{Ex4S}(=4.65\text{x}10^{-18}$ (cm^2 > 11:6 (5)

$_{Ex4P}(=1.17\text{x}10^{-17}$ (cm^2 > 13.2 (6)

$_i(=1.45\text{x}10^{-17}$ (cm^2 > 15.75 (7)

-Because of the lack of experimental data concerning the collisional ionization of excited states for argon, we have adopted the same assumption as made by Evans and Gamal (1984). Therefore, these cross sections are assumed to have the same analytical expressions as that of the ionization of the ground state atom with some adjustment for the numerical factor such as:

$_{i4S}(=1.45\text{x}10^{-15}$ [cm^2≥(15.75-11.6) (8)

$_{i4P}(=1.45\text{x}10^{-15}$ [cm^2 ≥(15.75-13.2) (9)

-Photo-ionization coefficients of the excited atoms are estimated from a formula given by Grey Morgan (1975) to be

$$C_q = \sigma^q/(q-1)\nu_L^{q-1}h^q \qquad (10)$$

where σ^q is the atomic cross section (typically≈ 10^{-16} cm^2), ν_L is the laser frequency, h is Plank's constant and q is the number of photons absorbed by an atom to be ionized.

Under the experimental conditions of Rosen and Weyl (1987) (λ= 0.53µm) two photons are required to photo-ionize both the 4S state and the 4P state. Therefore, the estimated photo-ionization coefficient using this formula is given by

$C^2 = 1{:}27 \times 10^{-14}\ s{-}1\ (W\ cm^{-2})^{-2}$ (11)

which is in accordance with that given in Weyl and Rosen (1985). In this case the photo-ionization of the 4P state results in an electron with an energy ≈2 eV.

-The three-body recombination coefficient is taken from Weyl and Rosen (1985) as

$R_{3b}=5.6 \times 10^{-26} n/\ \ ^{9/2}\ cm^3\ s^{-1}$ (12)

where n is the electron density.

-Finally, the electron diffusion coefficient is calculated using the same equation as that used by Rosen and Weyl (1987), namely

$$v_D \frac{2\varepsilon}{3m v_C}\frac{1}{\Lambda^2} \qquad (13)$$

where Λ is the diffusion length and as in the present case when the cylindrical focal volume of radius b is considered, Λ is approximated by Λ= b/2.405 and is the average electron energy. Ambi-polar diffusion is ignored in this work as it is insignificant under the experimental conditions considered in this analysis (Weyl and Rosen, 1982).

The temporal variation of the laser intensity is taken as Gaussian shape and the focal volume is considered to be cylindrical with radius r and axial length z. .The breakdown criterion adopted in this work is the attainment of ionization 0 .1% of the atoms present in the focal volume. The spatial distribution of the laser intensity is considered to be varied with the length of the focal volume such as:

$W(z)= W_0\ (1+z^2/z^2_R)^{1/2}$ (14)

Where z_R is the Rayleigh length.

Considering these cross sections and rate coefficients a computer program was undertaken to investigate experiments on the breakdown of argon by laser radiation.

3. Results and Discussion

Applying the considered model computations are conducted to obtain the threshold intensity for breakdown and plasma development as a function of the gas pressure. Comparison between these values and those experimentally measured by Bindhu *et al.* (2003) is shown in figure (1).Good agreement is obtained where both showed decrease of the threshold intensity/laser input energy as the gas pressure increases. This result confirms the validity of the model to investigate the experimental measurements considered in this study. In order to study the physical processes responsible for breakdown as a function of gas pressure figure (2) illustrates the EEDF calculated at the end of the pulse for different values of the gas pressure.

This figure showed that as the gas pressure increases the EEDF increases with its tail directed towards the energy range which is almost coincide with the ionization limits.

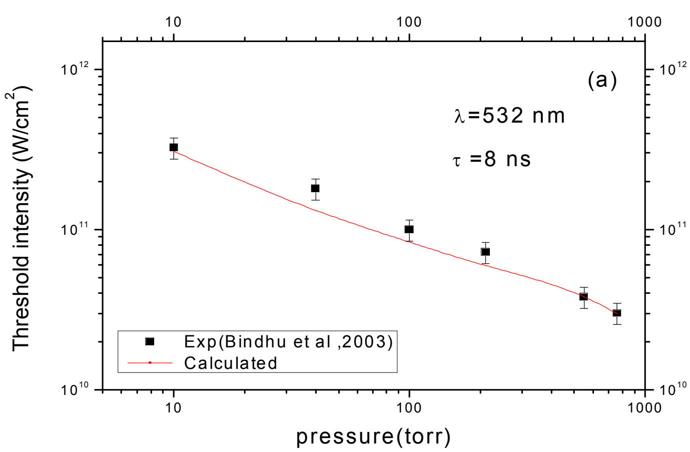

Fig.1: Comparison between the calculated and measured threshold intensities as a function of gas pressure

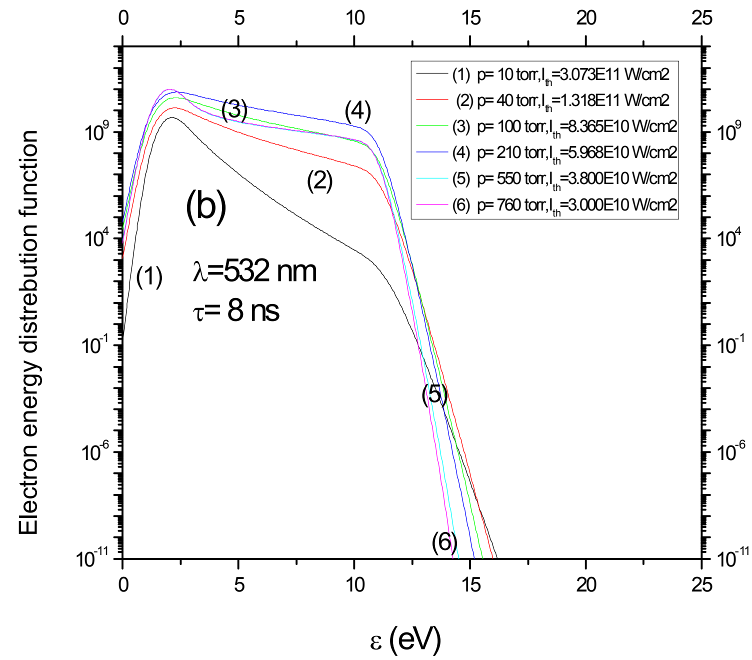

Fig.2. The EEDF calculated at the end of the laser pulse for different values of gas pressure.

To assure the correlation between the physical processes and gas pressure, figure (3) represents the time evolution of the electron density at the different pressure values. It is clear from this figure that at the low pressure value the electron density increases slowly during the early stages of the laser pulse .This is attributed to high competition rate between the generation of electros through photo-ionization process and its losses from the vocal volume by diffusion at this pressure range. As the gas pressure increases collision processes may contribute pronouncedly to the electron growth rate beside the photo-ionization process. This in turn illustrate the fast increase of the electron density near the peak of the laser pulse shown by curves(5) and(6) in this figure.

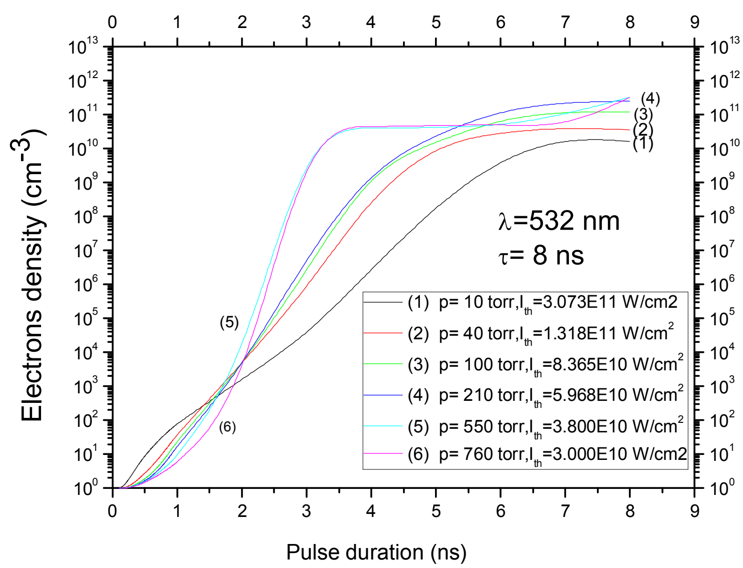

Fig.3. Time evolution of the electron density at the different values of the gas pressure.

In figure (4) the time variation of the electron mean energy is presented for the different pressure values. This figure illustrates the role played by the photo ionization process at the low pressure value , where the electron mean energy starts with high value(4 eV) then it decrease fast down to a value of 2 eV ,where it continue at this value up to the end of the pulse This confirms the fact that ionization at this pressure proceeds via photo ionization processes . At the intermediate pressures ,although the electron mean energy starts at the same value ,but it undergoes a fast decrease followed by a noticeable increase at the end of the pulse .At high pressures different behaviour is observed where the electron mean energy suffers from an increase around the peak of the pulse ,this clarifies the role of gain processes which could easily overcome the loss processes.

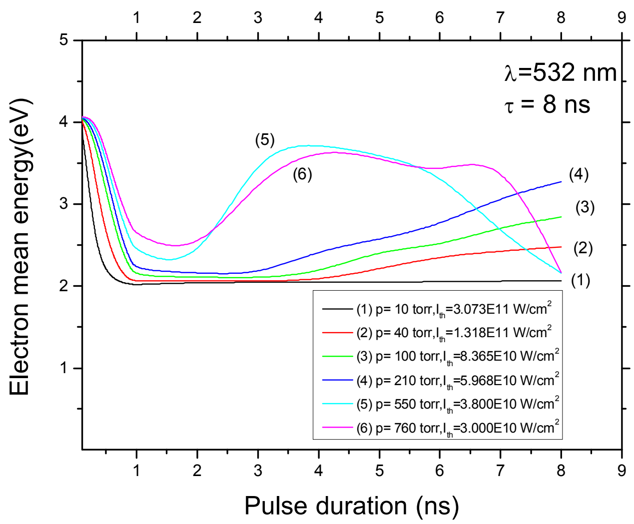

Fig.4: Electron mean energy plotted against time for the different values of the gas pressure.

To study the effect of the physical processes on the characteristics of the formed plasma ,figure (5) illustrates the contour image of the electron energy distribution zones that represent the formed plasma at gas pressure value 210 torr. This value is deliberately selected since it represent the most lengthy formed plasma among those calculated at different pressures.

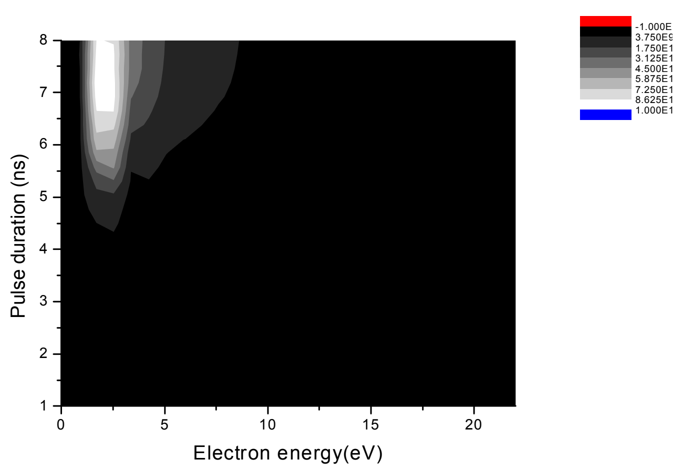

Fig.5 : Contour lines of the calculated temporal variation of the Electron energy distribution zones at pressure 210 torr.

To study the effect of gas pressure on the plasma propagation in the focal volume ,calculations are carried out to find out first a relation between the absorbed and scattered energy as well as the threshold intensity as a function of gas pressure covering a range 1.0 -100 atm (10 – 760 t0rr) at laser energy corresponds to the breakdown condition 230 mJ, (Yamada *et al.,*1994). This is shown in figure (6) ,where the absorbed energy reaches its maximum value at 760 torr. This indicates that the absorbed energy may exhaust in plasma expansion in the breakdown region under this experimental conditions.

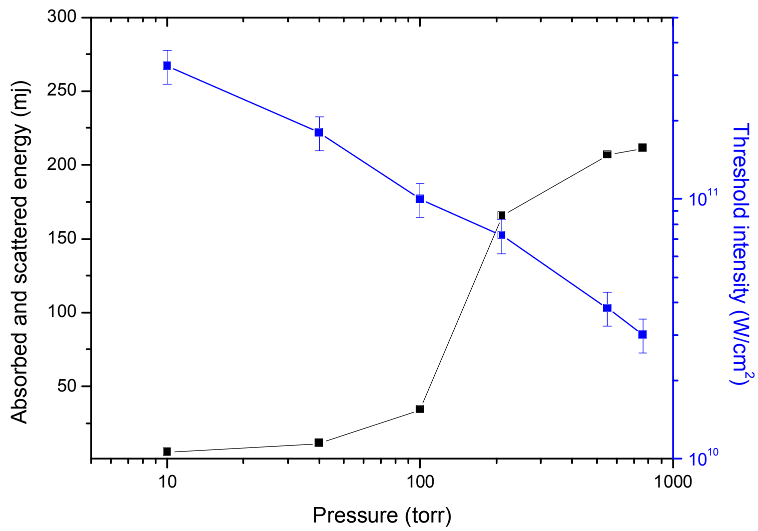

Fig. 6 Relation between the absorbed and scattered energy and the corresponding values of the threshold intensities plotted as a function of gas pressure.

Figure (7) shows the variation of the intensity as a function of the input energy at different axial points along the axial distance of the focal volume at laser input energies 12 mJ , 55 mJ and 155 mJ. It is noticed here that at the highest energy the plasma expands to a distance lies between 0 and z_R (The central point and the Rayleigh length). This means that as the input energy increases the plasma propagates more towards the laser beam.

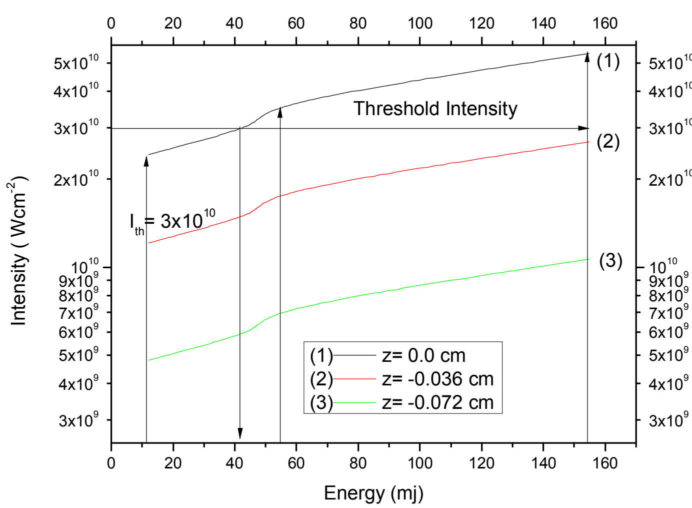

Fig.7. Variation of the intensity as a function of the laser input energy at different values along the axial distance.

To confirm this result a relation between the electron number along the axial distance at laser powers 7 MW and 20 MW is plotted in figures (8 ,9) to specify the actual axial distance at which breakdown occurs , this in turn identifies the length of the formed plasma .Increasing the laser power results in an increase of the plasma length despite the value of the gas pressure .

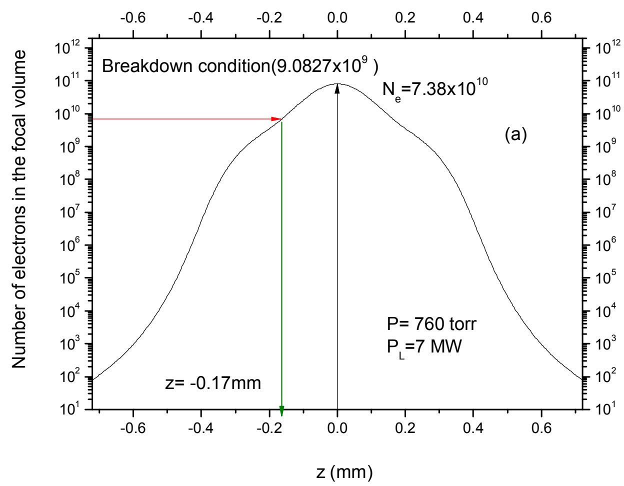

Fig.8 Variation of electron number as a function of both the axial and radial distances at input power 7 MW

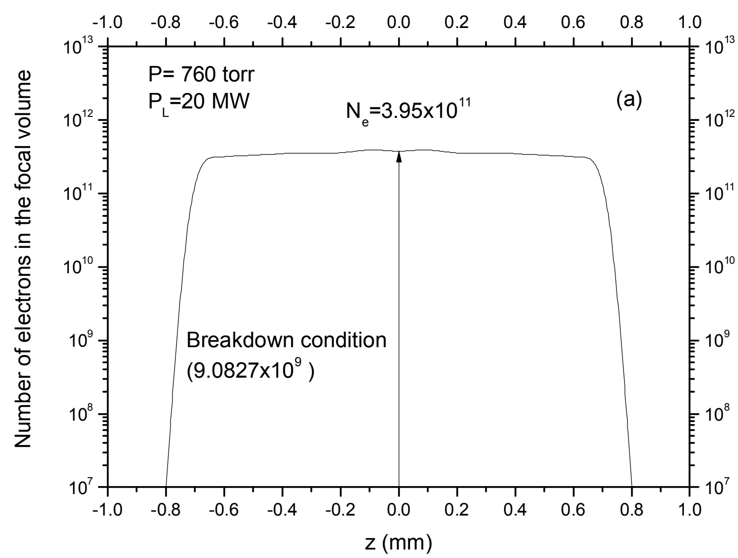

Fig. 9 The same as in figure 8 but at laser input energy 20 MW.

4. Conclusion

The electron cascade model presented in this work provided a reasonable interpretation on the effect of gas pressure on the physical processes responsible for the breakdown of argon over a pressure range 0.013-100 atm (10 – 760 torr) by the second harmonic of a Nd:YAG laser source with 8 ns pulse duration. This is confirmed from the good agreement which obtained between the calculated threshold intensity as a function of the gas pressure and the experimentally measured ones. The calculation of the EEDF and its parameters underlined the characteristics of the formed plasma in the breakdown region and its relation with the gas pressure. Electron diffusion acts to deplete the electron density at the low pressure regime. More over the study of the spatial and temporal variation of the laser intensity in the focal volume showed the exact correlation between laser input energy ,gas pressure and plasma expansion and propagation along the axial distance.

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for 0.3 < < 20 eV