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INTERNATIONAL CENTRE FOR
THEORETICAL PHYSICS

LAMP SERIES REPORT

(Laser, Atomic and Molecular Physics)

INVESTIGATIONS OF BREAKDOWN OF RARE GASES BY SHORT PULSES OF 1.06 μm LASER RADIATION

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INTERNATIONAL ATOMIC ENERGY AGENCY

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MIRAMARE-TRIESTE

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INVESTIGATIONS OF BREAKDOWN OF RARE GASES BY SHORT PULSES OF 1.06 μm LASER RADIATION

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ABSTRACT

An analysis based on a numerical integration of the equation which governs the total growth rate of the electron density due to the combined effect of multiphoton ionization and electron cascade ionization processes is presented. The calculations are carried out for argon and xenon gases in the pressure range $1.0 - 5 \times 10^3$ Torr illuminated with $10ps$ pulses of $1.06\mu m$ laser radiation. This analysis shows that the two ionization processes (multiphoton and electron cascade) play an important role over the whole pressure regime considered. At low pressures (< 300 Torr) the breakdown is governed solely by the multiphoton process, indicated by the weak dependence of the threshold intensity on the gas pressure. At higher pressures (< 760 Torr) however, a strong pressure dependence of the threshold intensity is observed. This is considered as evidence for the domination of the cascade ionization process at these pressures. On the pressure range 200 – 760 Torr a slight decrease of the threshold intensity is observed for both gases. This is attributed to the minor role played by the cascade ionization process together with the multiphoton ionization process at these pressures. The calculated thresholds for argon and xenon are consistent with those experimentally measured.

1. Introduction

The pressure dependence of the threshold intensity, I_{th}, of breakdown of gases by brief pulses of laser radiation has been of interest to many workers (Alcock and Richardson 1968, Aaron et al 1974, Ireland 1974, Dewhurst 1977) in attempting to determine the fundamental energy coupling mechanisms responsible for the breakdown phenomenon. It is generally agreed that two main mechanisms are responsible for the almost instantaneous transformation of the gases into highly conducting plasmas. These mechanisms are: direct multiphoton ionization (MPI) and cascade ionization (CI). The relative importance of these two processes is shown to change according to the product of the gas pressure, p, and pulse duration, τ, (Grey Morgan 1975).

However, there is a measure of controversy in the literature between experimental measurements and the theoretical predictions, particularly in the investigation of breakdown of rare gases by brief pulses of Nd^{3+} laser radiation. This contradiction was observed from the strong pressure dependence of the breakdown threshold intensity, at pressures << 760 Torr, obtained in some experiments (Dewhurst 1977, Ireland and Grey Morgan 1974) where the breakdown was observed visually. This is not in agreement with the very weak pressure dependence predicted theoretically when MPI alone is taken to be responsible for the breakdown (Bunkin and Prokhorov 1967, Grey Morgan 1975).

Recently, measurements on the pressure dependence of I_{th} for argon and xenon at pressures 1.0 - 760 Torr illuminated by single 10 ps pulses of Nd^{3+} laser radiation (λ = 1.06 μm) are carried out by Avery (1984) in which a photomultiplier was used to detect the onset of breakdown. His results showed a very weak pressure dependence of I_{th} for p << 760 Torr. This weak dependence indicates the action of MPI process over this pressure regime which in turn contrasts sharply with the strong pressure dependence observed in the earlier experiments.

In this work a numerical analysis based on the integration of the continuity equation which governs the combined action of MPI and CI processes (Gamal et al 1981) is developed. This analysis aimed to study the exact correlation between the gas pressure and ionizing mechanism in the break-down phenomenon associated with the interaction of brief pulses of Nd^{3+} laser with argon and xenon gases under the experimental condition of Avery (1984).

2. Method of Calculation

The evolution of the electron density, n, due to the combined effects of MPI and CI processes is presented by the following equation

$$\frac{dn}{dt} = NW \ I^{k}(t) + n \nu_i \qquad (1)$$

The first term on the R.H.S. represents the MPI process where N is the density of neutral atoms in the gas, I(t) is the laser intensity in Wcm^{-2}, K is the number of photons required for ionizing the gas atoms and W is the MPI rate constant expressed in $(Wcm^{-2})^{-K}\ s^{-1}$. The second term of equation (1) describes the CI process where ν_i is the collisional ionization rate .given by (Ireland and Grey Morgan 1973).

$$\nu_i = N \left[\frac{377}{w^2} q \left(\frac{\nu_m}{N} \right)^2 \right] I(t) \qquad (2)$$

here q is a constant of a particular gas (typically of the order $10^{21}\ cm^{-1}\ s^{-1}\ V^{-2}$ for rare gases when I(t) is in Wcm^{-2}), w is the angular frequency of the laser radiation field and ν_m is the electron-atom collision frequency for momentum transfer which is assumed to be independent of electron energy (Grey Morgan 1975). The temporal behaviour of the laser pulse used in this analysis is assumed to be a triangular shape of duration 2τ such as :

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

$$I(t) = I_o\ (2 - t/\tau) \qquad \text{for} \qquad \tau < t < 2\tau$$

where I_o is the peak intensity of the laser flash.

A very simplified analytical solution of equation (1) based on the pressure regime assumption is given by Ireland and Grey Morgan (1973). In this work, however, a numerical solution of equation (1) using Runge-Kutta fourth order technique is presented. This enables us to solve the pressure regime problem where step by step integration of this equation is developed over the whole pressure range examined. Diffusion and recombination losses had not been taken into account since the characteristic times for these processes (~ nanoseconds, Ireland and Grey Morgan, 1973) are longer than the laser pulse length used in these calculations. The initial conditions used in solving equation (1) are n = 0 at t = 0, where the second electrons will be created by the MPI process.

3. Results

The analysis is used to interpret the experimental measurements of the I_{th} dependence on p , carried out by Avery (1984) for the case of argon and xenon gases irradiated with 10 ps pulses of Nd^{3+} laser radiation.

3.1 Argon

The results of computations are presented as a set of curves for the electron density, n, as a function of time for argon at pressure ranging from $1.0 - 5.0 \times 10^3$ Torr irradiated with a 10 ps pulses of Nd^{3+} laser radiation of peak intensity varies from 10^{12} to 10^{13} Wcm^{-2}. These curves are shown in figure 1 with the corresponding values of the gas pressure. In obtaining these curves the following parameters are used in the computations; $q = 1.0^{21}$ cm^{-1} s^{-1} V^{-2} , $\nu_m = 3.9 \times 10^9$ p s^{-1} Torr (Morgam et al 1971), K = 10 and $W = 8.0 \times 10^{-124 \pm 2}$ (Agostini et al 1970).

As we can see from this figure the growth of the electron density at all values of gas pressure is not instantaneous but it is considerably rapid up to the peak of the laser flash. Immediately after the peak two distinguishable behaviours of the electron density are observed: at low pressures (< 300 Torr, Curves 1-3) almost constant values of n are obtained during the second half of the laser pulse. At higher pressures (> 300 Torr Curves 4-7), however, the electron density continues to increase gradually ending with reasonably higher values at the late stages of the pulse duration. This figure also shows that curves (1-3) increase linearly with the gas pressure over the whole length of the laser flash. This behaviour was not observed at higher pressures (curves 4-7)

where the rate of growth of the electron density exhibits a considerably slow increase with the gas pressure during the first half of the laser pulse (notice the almost coincidence of the values of n for curves 3 and 4) followed by a faster increase during its second half.

From this figure we can infer that the growth of the electron density proceeds via two distinguishable mechanisms which are acting on different pressure regimes.

Considering the breakdown criterion as $\delta \sim 0.1$ % , where δ is the degree of fractional ionization of gas atoms in the focal region, curves (1-7) in figure 1 enables us to determine the time at which breakdown occurs, τ_B for each value of gas pressure. From the variation of the laser pulse intensity with the time and the determined value of τ_B at each value of gas pressure, we can obtain the corresponding value of the threshold intensity I_{th}.

Figure 2 shows the calculated values of I_{th} versus gas pressure. For an easy comparison, the experimental values of I_{th} (Avery 1984) are also shown in this figure. As we can see, our numerical results are in a reasonable agreement with the experimentally measured threshold intensities. The very weak dependence of I_{th} on the gas pressure observed at pressures $\leq$ 300 Torr gives evidence for the important role played by the MPI process on the ionization growth rate at these pressures. This result confirms the linear growth rate of the electron density with the gas pressure over the whole pulse duration observed by curves (1-3) in figure 1. A close study of figure 2 showed that over the pressure range 300 -760 Torr, the threshold intensities undergo a slight decrease. This decrease may be attributed to the minor role played by the collisional ionization process on the ionization growth rates together with the MPI process over this pressure regime. This result may in turn investigate the noticeable change of the behaviour of the electron growth (curves 4-6) shown in figure 1. Although there is no measured values of I_{th} at pressures > 760 Torr, however the calculated values showed a strong pressure dependence. The latter result indicates the domination of the CI process at these high pressures.

Concluding this point, we may remark that the growth of the electron density over pressure ranges $10 < p < 5.0 \times 10^3$ Torr takes place under the combined action of MPI and CI processes.

3.2 Xenon

Figure 3 illustrates the electron density versus time for the case of xenon over the pressure range $1.0 - 5.0 \times 10^3$ Torr (Curves 1-7), irradiated with 10 ps pulses of Nd^{3+} laser radiation with peak intensities varies as 10^{12} to 10^{13} Wcm^{-2}. The curves are calculated for the following parameters ; $q = 1.5 \times 10^{21}$ cm^{-1} s^{-1} V^{-2} (Ireland and Grey Morgan 1973), $\nu_m = 5.9 \times 10^9$ p s^{-1} Torr (Brown 1966), and $K = 9$, $A = 2.0 \times 10^{-112}$ (Agostini et al 1970).

From this figure, we can see that the behaviour of the growth of the electron density does not show any particular difference than those obtained for the case of argon (figure 1). However, an earlier deviation from linear growth for the electron density is observed in this case (curve 4 for P = 200 Torr). We also notice that curves (1-3) show an almost constant growth rate as the gas pressure increases over the whole pulse duration. This

behaviour was not observed for curves (4 and 5) but rather a slow growth rate is obtained on the first half of the laser flash followed by a faster growth on its second half. (notice the slight drop of the electron density values obtained for curve 4 at P = 400 Torr). Curves (6 and 7) in this figure show a considerable increase of the electron density during the second half of the laser pulse. This behaviour confirms the action of an ionizing process which has a fast growth rate at these pressures.

Following the same procedure used for the case of argon, curves (1-7) in figure 2 enable us to compute the threshold intensities at each value of the gas pressure.

Figure 4 gives a graphic comparison of the computed values of the threshold intensities and the measured ones (Avery 1984). From this figure it can be seen that a reasonable agreement is obtained between the calculated and measured threshold intensities over the whole range of the gas pressure examined experimentally. The weak pressure dependence of I_{th} observed on the low pressure range (10 - 200 Torr) depicts the main contribution of the MPI process at these pressures. This result may also describe the linear growth of the electron density with the gas pressure shown by curves (1-3) in figure 3. Another point of interest to examine is the strong pressure dependence of I_{th} observed in figure 4 at pressures > 760 Torr. As we can see the I_{th} dependence on p is such as $I_{th} \propto p^{-1}$ which gives an evidence for the domination of the CI process at high pressures. Again, the gradual decrease of the threshold intensity observed over the pressure range 200 < p > 760 Torr can be understood as a result of the small influence of the CI process on the electron growth rate at these pressures.

From the above results we may remark that the breakdown of xenon is governed by the combined action of the two ionizing processes MPI and CI.

4. Conclusion

We have calculated the threshold intensity for breakdown of argon and xenon at pressures varying from 10 —— 5×10^3 Torr irradiated with 10 ps pulses of Nd^{3+} laser radiation ($\lambda = 1.06\ \mu m$). The dynamic dependence of I_{th} on p for the two gases showed that the breakdown mechanisms for both of them are of a similar nature acting on different pressure regimes. At low pressure the MPI process is dominant. As the pressure increases the effect of MPI start to decrease while CI process begins to give a small contribution to the ionization rate up to the atmospheric pressure (760 Torr) where the domination of the CI process is predicted.

No sharp transition pressure is observed instead there is a region in which both processes are contributing to the breakdown phenomenon. This region depends mainly on the type of the gas (Ionization potential, momentum transfer collision frequency...etc.) and the characteristics of the laser used in the experiment (pulse duration and wavelength).

This conclusion indicates the important role played by the MPI and CI processes in obtaining the instantaneous transformation of the gases into plasma. Moreover this result shows the significance of MPI process at low pressure observed in this recent experiment.

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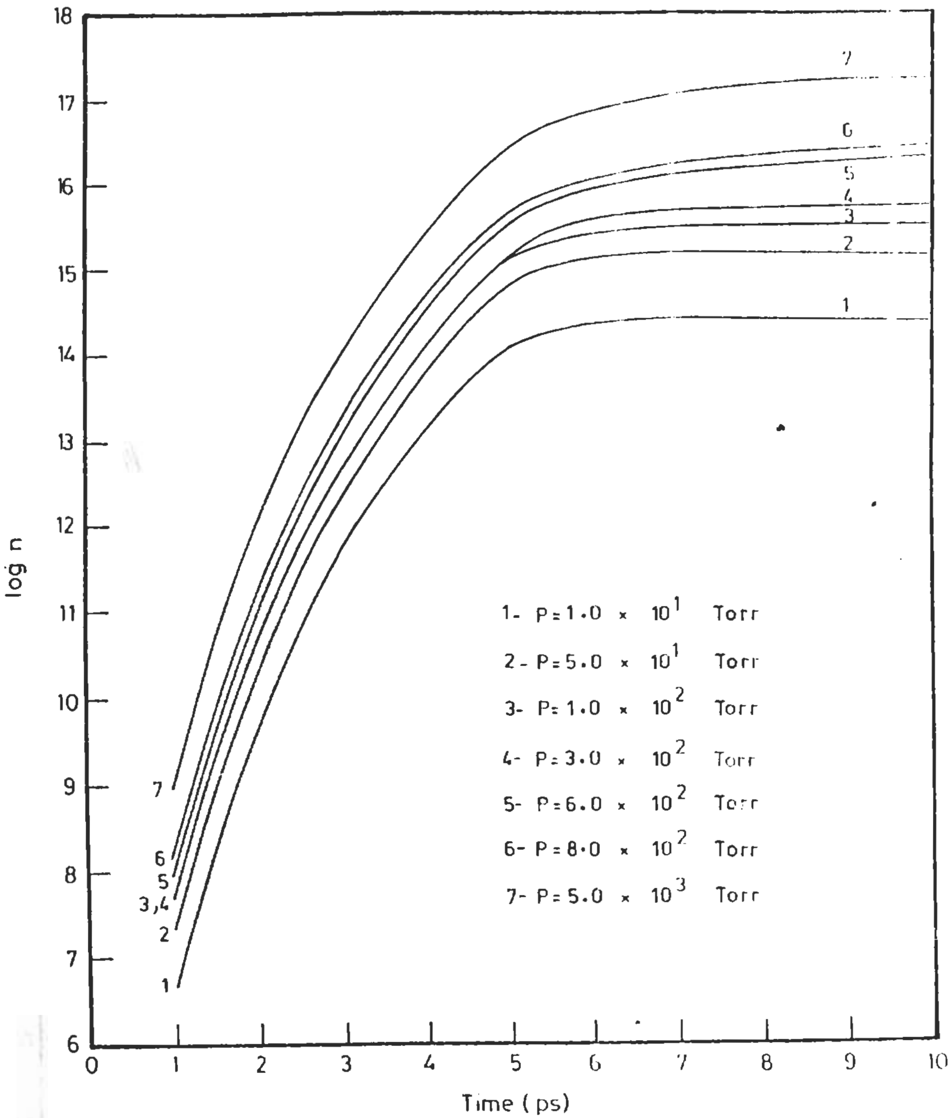

Fig.1

The evolution of the electron density during the laser flash for argon at different values of gas pressure irradiated with 10 ps pulses of Nd^{3+} laser radiation.

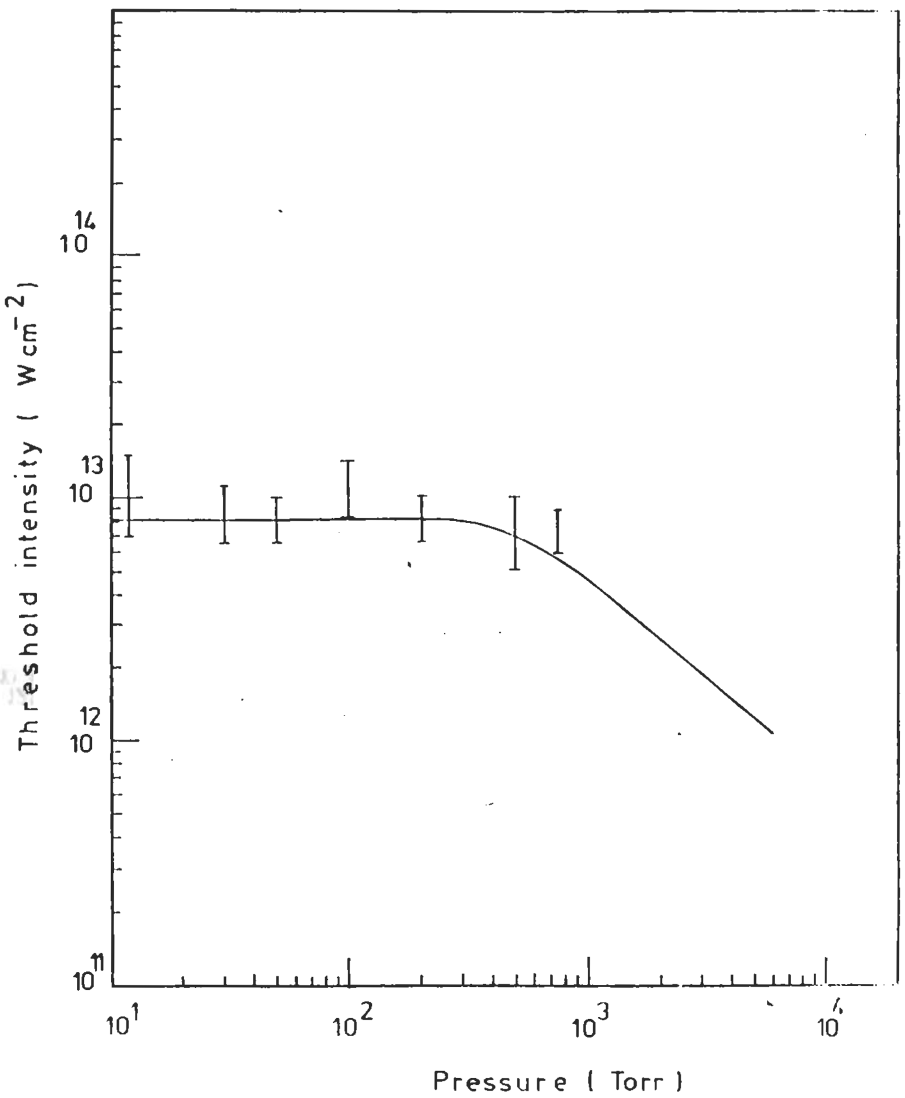

Fig.2

Comparison between the calculated threshold intensities of argon at different pressures (solid line) and the experimentally measured ones (1).

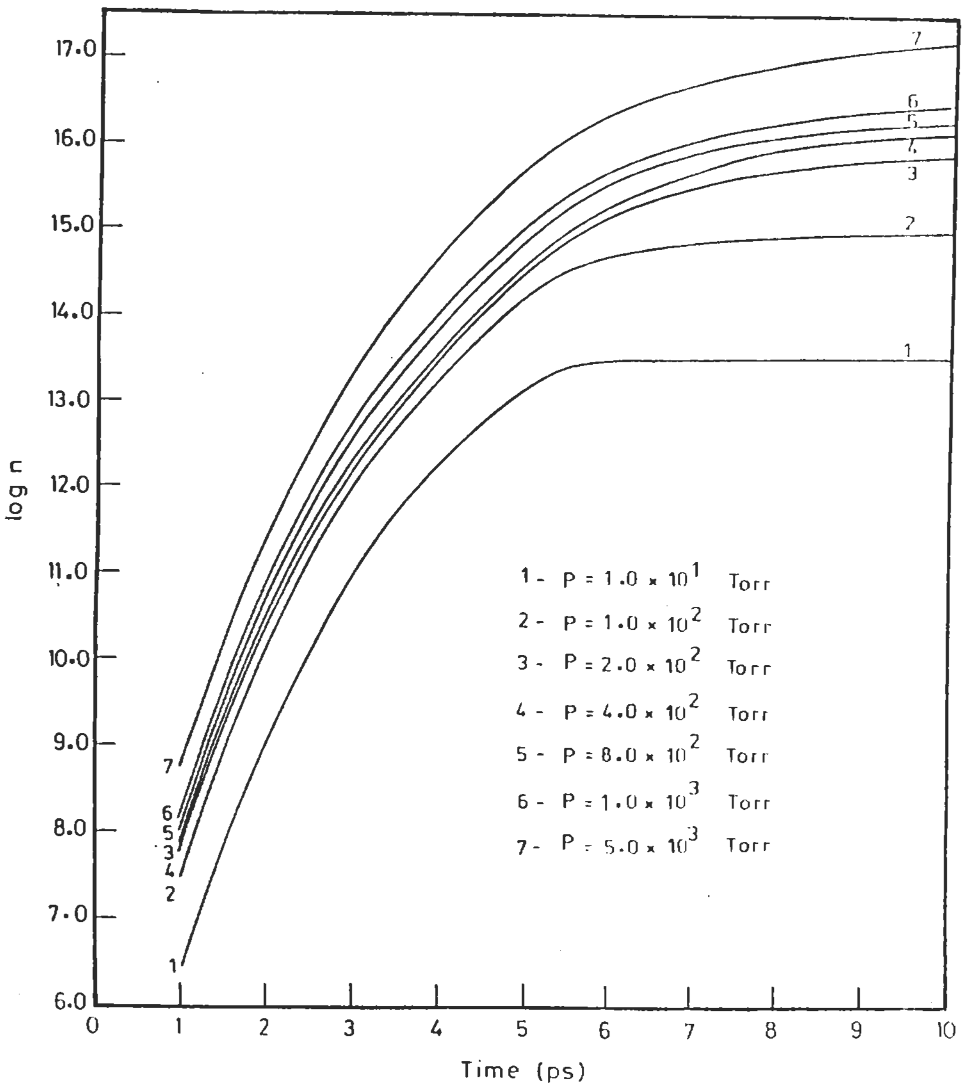

Fig.3

The same as in figure 1 but for the case of xenon.

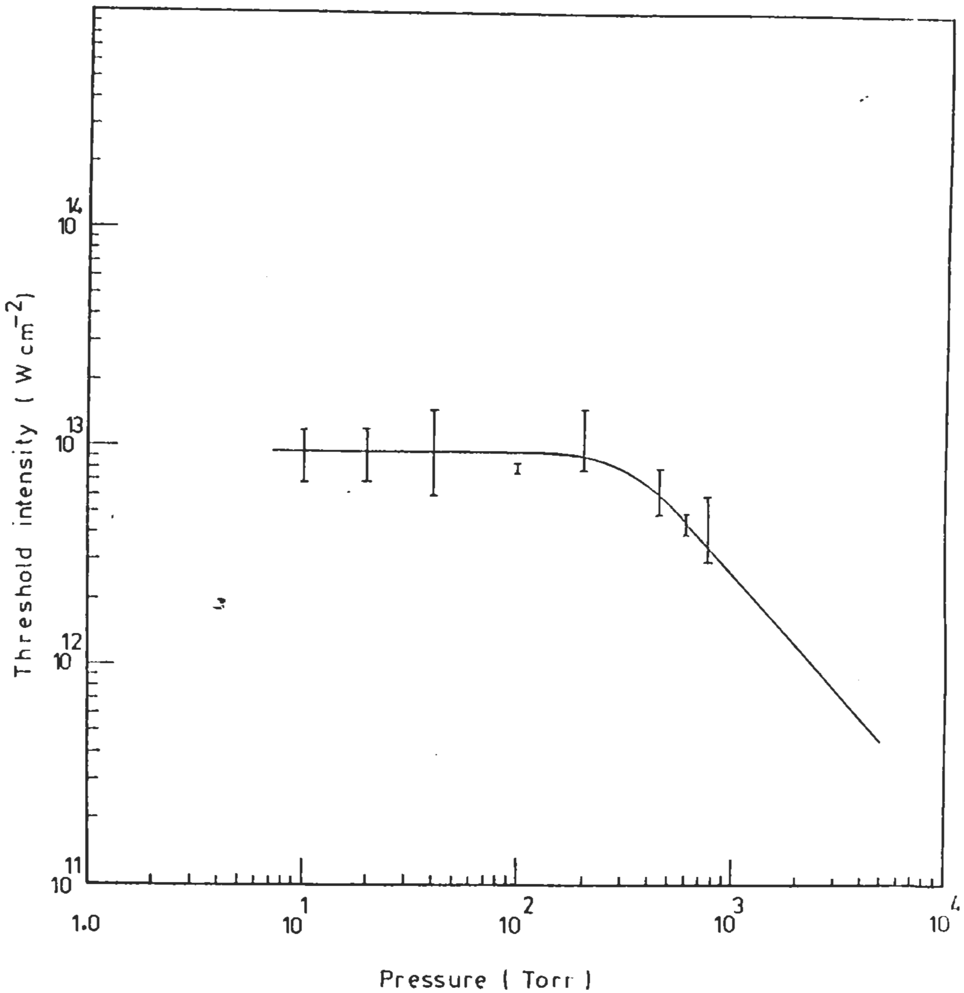

Fig.4

Threshold intensities as a function of gas pressure for xenon. Calculated values (solid line), experimental values (I).