Classification
Physics Abstracts
42.60B — 42.55H

Observations on the power of a N₂ laser

B. Giannetas, P. Persephonis and R. Rigopoulos

Département de Physique, Université de Patras, Patras, Greece

(Reçu le 8 février 1985, révisé le 9 mai, accepté le 6 juin 1985)

Résumé. — L'étude de la puissance laser maximale en fonction de la pression d'azote a permis l'observation d'irrégularités dans certaines zones de pression. Ces irrégularités ne peuvent être expliquées par aucune théorie existante. Les auteurs les attribuent aux réactions entre les différents types d'ions présents dans le plasma d'azote. Les réactions $N_2 + N_2^+ \rightleftharpoons N_4^+$ et $N_2(M) + M_2(M') \rightarrow N_4^+ + e$ (ou) $N_2^+ + N_2 + e$ sont responsables des « traits » centrés vers 65 et 110 torr respectivement.

Abstract. — On the peak power output of a N₂ laser some « features » which appear as irregularities at certain pressure regions were observed. These cannot be explained by any of the existing theories. The authors attribute these « features » to reactions among ion types in the N₂ plasma. The reactions $N_2 + N_2^+ \rightleftharpoons N_4^+$ and $N_2(M) + M_2(M') \rightarrow N_4^+ + e$ (or) $N_2^+ + N_2 + e$ are the causes of the « features » centered in about 65 and 110 torr respectively.

1. Introduction.

A gas laser is an integrated electrooptical system. Its laser action is due to an electric discharge in a tube full of a specific gas.

By the electric discharge the population inversion between two energy levels of the laser gas necessary for the laser action is achieved.

Especially for the pulsed lasers, as is the N₂ laser, the population inversion is achieved by an, as much as possible, abrupt appearance of potential difference between two electrodes in the laser tube. This results in an abrupt increase of the corresponding current.

The above can be realized by an appropriate electric circuit. For the T.E. N₂ lasers the « Blumlein circuit » is widely used. This circuit is consisted :

(a) of the transmission lines (which also serve as capacitors) and (b) of the ignition system (i.e. spark-gap).

A special concern is given so as to have the smallest possible inductance from the several elements of the circuit and the ignition system in particular.

In a previous work [1, 2] a method was developed to increase both the peak power and energy per pulse in a N₂ laser. By this method, a quadruple and then a sixfold line were used, instead of the usual double transmission line.

The importance of the discharge preionization on the laser efficiency has also been demonstrated. The above

results are in agreement with those of other authors [3, 4] and may be recapitulated as follows :

The statistical time lag depends on the preionization. The bigger the statistical time lag, the higher the actual starting voltage and consequently the higher the peak power and the energy per pulse.

It is also known that the ions remaining in the laser tube from a discharge play the preionization role for the next discharge.

2. Experimental results.

In this work a double transmission line N₂ laser was constructed as shown in figure 1. Its transmission line can also be replaced with a quadruple one as is described in [1]. The above are shown in figure 2.

By using the above arrangement the peak power *versus* the laser tube pressure was examined. The results of those measurements are grouped as follows :

2.1 Measurements with an uncontrolled but self adapted flow of the gas, that is measurements taken by adjusting the inlet vane of the gas only, while the outlet vane aperture remained unchanged. Thus by increasing the gas pressure, the flow of the gas increases too. As it is shown in figure 3 the flow *versus* the pressure increases linearly. Thus a pressure region from 35 to 130 torr was covered while the flow increased from 3.4 lt/min (35 torr) to 4.8 lt/min (130 torr).

Fig. 1. — Experimental set-up of the N₂ laser and the measurement system of the light output.

Fig. 2. — Schematic drawing of the double (a) and quadruple (b) parallel plate transmission lines nitrogen laser.

The results of the measurements taken with self adapted flow, are shown in figure 4. In this figure the peak power *versus* pressure for different voltages is plotted (repetition rate 10 Hz).

2.2 Measurements with constant flow of the gas, that is measurements taken by adjusting both inlet and outlet vanes simultaneously so that the flow remained constant and equal to 3.4 lt/min while the pressure changed from 35 torr to 130 torr.

The results of the measurements taken with constant flow are shown in figure 5. In this figure the peak

power *versus* pressure for different voltages is plotted (repetition rate 10 Hz).

As one could see in figures 4 and 5, the curves of the peak power *versus* pressure exhibit « features » which appear as irregularities in the peak power curves at some pressure regions (about 65 torr and 110 torr). These cannot be explained by any of the existing theories [5-6].

Measurements have been repeated many times using sparkgaps of different inductance. The results confirm the existence of the above « features ». The measurements were also repeated with a quadruple line N₂

Fig. 3. — The gas flow *versus* N₂ pressure for self adapted flow (a) and constant flow (b).

Fig. 4. — Laser peak power *versus* laser tube pressure, for self adapted gas flow in a double transmission line N₂ laser (repetition rate 10 Hz).

Fig. 5. — Laser peak power *versus* laser tube pressure for constant gas flow in a double transmission line N₂ laser (repetition rate 10 Hz).

laser [1, 2]. These are plotted in figures 6 and 7 for self adapted flow and constant flow respectively. As it is shown in figures 6 and 7 similar « features » characterize the peak power *versus* pressure plots and in the same pressure regions as in the double line cases.

It is worth to be emphasized that all the above measurements showed in figures 4-7, have been taken with a lot of care. The points' size in figures is an indication of the experimental errors. So it is sure that the observed « features » are physical effects.

Fig. 6. — Laser peak power *versus* laser tube pressure, for self adapted gas flow in a quadruple transmission line N₂ laser (repetition rate 10 Hz).

Fig. 7. — Laser peak power *versus* laser tube pressure for constant gas flow in a quadruple transmission line N₂ laser (repetition rate 10 Hz).

The above « features » are more intense in the constant flow case. This supports the hypothesis that these « features » may be attributed to corresponding variations of preionization.

Actually the conditions in which the two groups of data were taken, as described above, differ only in the ion population in the laser tube (all the other conditions remain constants). This population is larger in the constant flow case than in the corresponding self adapted one for the same pressure.

This is because as it can be seen in figure 3 the gas in the constant flow case remains in the tube for a larger time. Consequently the ion population difference (that is the preionization difference) is the main factor for the differences in the peak power between the measurements groups.

If the cause of these « features » is really the pre-ionization the actual starting voltage which depends on the preionization [3, 4] will have to show corresponding « features ».

The actual starting voltage was measured *versus* pressure and the E/p was calculated (Fig. 8). A high voltage probe (Tectronix P 6015) was used. Actually in these measurements irregularities in the same pressure regions were observed as it is in the case of the peak power curves. Consequently the « features » in the peak power correspond to variations of pre-ionization.

Fig. 8. — Actual starting voltage (V_{act}), ratio E/p and laser peak power ($P.p.$) *versus* laser tube pressure for constant gas flow and double line N_2 laser (Appl. Volt. 12 kV repetition rate 10 Hz).

It seems [4, 8] that many authors have already measured similar « features » in different pressure conditions. In general, however, they disregarded them in fitting their curves. Figures 9, 10 and 11 have been taken indicatively from the works of these authors [4, 8]. In these figures we have fitted also their results with curves similar to the ones of figures 4-7.

3. Nitrogen ions and ion reactions.

An electric discharge starts when a potential difference appears across the electrodes in the laser tube. The potential difference begins to appear when the spark-gap is fired and increases very quickly with the time.

The residual positive ions from the previous discharge play the preionization role, as mentioned above. Thus during the potential difference increase between the electrodes, the residual ions move towards the cathode. The ions acquire energy from the electric field and they release primary electrons from the cathode surface. The discharge begins with the primary electrons appearance. Then the increase of the potential difference stops. At this time the potential difference reaches its maximum value (actual starting voltage) and then begins to collapse [6, 7].

Fig. 9. — Enhancement of the laser output at 357,7 nm by addition of C_3H_8 gas (V_a : ○, 15 kV; ●, 18 kV; △, 21 kV; ▲, 24 kV): $P(N_2) = 20$ torr; $\nu_r = 1/3$ Hz. Tetsuya Mitani [4]. (----- our plotting).

Fig. 10. — Output at 3 577 Å as a function of total pressure with partial pressure of N_2 as parameter (○, 25 torr; △, 50 torr; □, 75 torr). Junichi Itani [8]. (----- our plotting).

As it was mentioned in the previous section, the « features » in the peak power correspond to variations of preionization. But except their great number, the ions in the tube are generally of different type.

Fig. 11. — Output at 3 371 Å as a function of total pressure with partial pressure of N₂ as parameter (O, 25 torr; Δ, 50 torr; □, 75 torr). Dashed line indicates the output at 3 371 Å in the case of N₂ gas alone. Junichi Itani [8].

Ions of different type have different mobilities and they require different statistical time to reach the cathode and release primary electrons [9]. However, the actual starting voltage is proportional to this statistical time. Consequently, ions with different mobilities form different actual starting voltages and finally the laser irradiates different optical output energy.

By changing the N₂ pressure in the laser tube the ion population is changed « normally », that is according to collisions theory and it does not cause particular variations in the peak power. On the other hand the ions type may be changed, through ionic reactions, under certain values of E/p . Conclusively by changing the N₂ pressure the only variation of preionization causing « abnormal » variation in the peak power, is the change of the ion type. Only through ionic reactions the change of the ion type could be achieved. In certain pressure regions these ion reactions are favoured.

Briefly, the following are known about the ions and ionic reactions of the N₂ [9-25]: there are four main ion types of the nitrogen. N⁺, N₂⁺, N₃⁺ and N₄⁺ [9-11]. The appearance potentials and the zero field mobilities are given in table I [11-13].

When the electric discharge starts, electrons flying towards the anode collide with N₂ molecules and form both the primary nitrogen ions N⁺, N₂⁺ and excited

Table I. — The observed appearance potentials and the zero field mobilities for ions in nitrogen.

Type of ions	N ⁺	N ₂ ⁺	N ₃ ⁺	N ₄ ⁺
Appearance potential (Volt)	24.2	15.5	22.1	15.8
Zero field mobility (cm ² /V.s)	2.97	1.87	2.26	2.33

molecules [12]. Both, the ions and excited molecules, under certain conditions of pressure and electron temperature T_e ($T_e \propto E/p$), further react with nitrogen molecules and form secondary ions N₃⁺ and N₄⁺ [12]. The primary ions N⁺ and N₂⁺ are formed according to the reactions:

and

It is known that charge exchange always occurs with maximum cross section in any gas for the ion which becomes the normal gas molecule. The inverse reactions have also maximum cross section. In this case the N₂⁺ is the most abundant of the primary ions [14].

For the N⁺, as far as this ion is concerned, there are not indications that it reacts with other molecules to form new ion types [10, 12, 13]. From the above is concluded that the N⁺ ion plays an unimportant role in the discharge.

The complex (secondary) ions N₄⁺ and N₃⁺ are formed by the N₂⁺ ion through ionic reactions. Thus many authors have shown the close relationship between the ions N₂⁺ and N₄⁺ through the famous three body reaction [10-21].

This reaction shows that a part of the N₂⁺ ion population is transformed in flight, into a faster ion, the N₄⁺.

This process was suggested as a hypothesis by Varney [14] when he observed an irregularity in the curve of drift velocities versus E/p_0 for ions in nitrogen.

Varney also suggested the application of the thermochemical equilibrium, theory, for the two way reaction [16, 17].

If ε is the N₄⁺ ions fraction at temperature T , then, the equilibrium constant K is given by the relation:

$$K = \varepsilon p / (1 - \varepsilon)$$

where p is the gas pressure. The Nerst equation then gives:

$$\ln K = -(\Delta H/RT) + R^{-1} \int \left\{ (C_{p2} + C_{p3} - C_{p1}) dT \right\} T^{-2} dT + \Delta S_0/R$$

where ΔH is the heat of dissociation C_{p1} , C_{p2} , C_{p3} are the respective molar heat capacities at constant pressure of N_4^+ , N_2^+ and N_2 and ΔS_0 is the difference between the molar entropy constant of N_4^+ and that of the dissociation products. R is the gas constant. The N_2^+ and N_4^+ ion mobilities versus E/p or E/N are shown in figure 12 [22].

Fig. 12. — Mobilities of nitrogen ions in nitrogen. — Mosely *et al.* — — — Mc Knight *et al.* — · — Saporochenko [22].

Experimental results [11] confirm that at low E/p (that is at high pressures) the two ions (N_2^+ , N_4^+) are quite distinct and the N_4^+ is much more abundant. A quite different situation is observed when E/p increases. Thus, for $E/p > 60 \text{ V.cm}^{-1}.\text{torr}^{-1}$, the mobility of the ions which corresponds to the one of N_4^+ (as observed by Saporoschenko) falls very rapidly and approaches the one of N_2^+ (Fig. 12). This means that a considerable fraction of N_4^+ ions has been converted into N_2^+ ions. As E/p further increases (that is the pressure decreases), the N_2^+ ion is the most abundant.

The reverse reaction (3) was observed also experimentally by M. Saporoschenko. The spectrum of these ions, as it is shown in figure 13 for different values of E/p_0 , has « features » that suggest the existence of reactions involving these ions.

The reaction (3) is taking place at electron energy greater than 17.5 eV.

At electron energies close to the appearance threshold of the N_2^+ and N_4^+ ions, a second indirect

Fig. 13. — Pulses of ion transients observed by Saporoschenko. ● N_4^+ , ○ N_2^+ , for the following values of E/p in $\text{V cm}^{-1} \text{ torr}^{-1}$: a) 44.3, (b) 60, (c) 67, (d) 101.5.

ionization mechanism occurs in the collision of vibrationally excited molecules or electronic metastable molecules [23-25]. The ionization processes are :

$$E(v) + E(v') \geq 15,6 \text{ eV}$$

$$E(v) + E(M) \geq 15,6 \text{ eV}$$

$$E(M) + E(M') \geq 15,6 \text{ eV}$$

H. Brunet *et al.* [25] came, both from theoretical and experimental results, to the conclusion that most of the ionization occurs through process III.

Finally the N_3^+ ion is formed by the reaction

This reaction may take place but the reaction rate is very small because of the eclectic use of the rare $N_2(^4\Sigma_u^+)$ ion.

On the other hand the N_3^+ and N_4^+ are rarely found together because very high ionizing electron energy is needed to produce the N_3^+ ions. Under these conditions the N_4^+ ions [9] are dissociated. So the fraction of the N_3^+ ions in the gas laser plasma is small and its role is nearly unimportant [10, 12].

4. Discussion.

In a N_2 laser the time interval between the start of the discharge and the lasing action is between few to 10 ns. During this short time interval, collisions between molecules and ions or ion recombinations can be ignored. Consequently ionization and excitation of the gas in this time interval is almost entirely due to collisions of molecules with the electrons. These processes are described by the reactions (1) and (2). Ionic reactions appear after the lasing action and they alter the population distribution of different types of ions and metastable molecules in the gas. These ions play a significant role in the next discharge, being the preionizing ions affecting the actual starting voltage.

The experimental results of this work show that not only the total number of ions but even the ion type existing before the discharge in the laser tube, affect the optical output of the laser.

The external parameters through which the preionization is altered are :

(a) The pressure, (b) the pulse repetition rate, (c) the flow of the gas and (d) the applied voltage.

Unfortunately these external parameters are very few and very poor knowledge exists of what exactly happens in the N₂ plasma. Consequently, the quantitative study of this effect is fairly difficult.

Qualitatively, bearing in mind the above it is possible to make some estimations : the results of the measurements with constant flow, for double line (Fig. 5) show that, for low pressures, the peak power increases up to 60 torr. A small fall of the peak power follows at about 65 torr and for higher pressures the peak power continues to increase until it obtains its maximum value.

The irregularity observed in the pressure region around 65 torr, is, as mentioned above, attributed to the significant alteration of the type of ions which dominate in the plasma. The predominant ion for $p < 65$ torr is different from the predominant ion for $p > 65$ torr. In the region around 65 torr the first ion is converted into the second one and *vice versa* through an ionic reaction which takes place in a relatively narrow region of E/p .

By extrapolating the peak power curve as shown in figure 8 one can conclude that the ion which dominates at low pressures « produces » more light intensity than the ion which dominates at higher pressures. As known for higher ion mobility the statistical time lag is lower and therefore is lower the actual starting voltage and, finally, the laser peak power. Consequently, in our case, the predominant ion in pressures $p > 65$ torr is quicker than the predominant ion in pressures $p < 65$ torr.

From the above is concluded that this irregularity observed in the peak power may be attributed to the ionic reaction (3) for three reasons :

(a) The N₂⁺ ion exists in abundance. Thus a large number of ions take part in the reaction. Consequently, the change of the ions character becomes significant and affects the laser efficiency (through the preionization). This irregularity in peak power is more intense than other ones observed.

(b) In this ionic reaction the predominant ion at high pressures, (N₄⁺), is quicker than the predominant one at low pressures, (N₂⁺), (Table I, Fig. 12). This is in agreement with the experimental results. It is therefore possible for the predominant ions in pressures $p > 65$ torr and $p < 65$ torr to be identified with N₄⁺ and N₂⁺ ions respectively.

(c) The ionic reaction (3) takes place at a small E/p region (Fig. 12) as it exactly happens in the ionic irregularity observed experimentally (Figs. 5, 7).

From figure 8 it is possible to estimate the degree of dissociation ε of the reaction

for every concrete value of pressure in the region where both ions exist ($\varepsilon = 1$ signifies complete disso-

ciation and $\varepsilon = 0$ no dissociation). Then the equilibrium constant K may be determined by the relation :

$$K = \varepsilon p / (1 - \varepsilon) .$$

This irregularity also appears, though less intensively, in measurements with constant flow in quadruple line (Fig. 7).

For the self adapted flow, the irregularity in the pressure region of around 65 torr takes a different form (Figs. 4, 6). Thus for the double line case (Fig. 4) the observed peak power decrease in constant flow measurements has already disappeared and the irregularity is displayed as a change in the rate of the peak power increase *versus* pressure. Yet, this irregularity appears less intense in the quadruple line case.

A second irregularity (however much weaker than the first one) is observed past the maximum of the power *versus* pressure curve. There the peak power decreases by increasing the pressure. This irregularity appears as a plateau in a large pressure region centred around 110 torr. It becomes stronger in quadruple line measurements and in particular for constant flow.

This irregularity may be explained as follows : in high pressures the electron temperature is low. This results in a significant decrease of the ions production, while the production of the excited molecules is favoured in metastable states and (or) vibrational levels of the ground state. What are important are in this case the reactions (5) because through them increases the ion population. Thus the peak power decrease *versus* pressure is hold and this appears in the form of plateaus.

5. Conclusion.

The significant rearrangements of the ion population performed through ionic reactions in a N₂ laser tube may be affected the laser output energy. The two main irregularities in the peak power *versus* pressure curves observed experimentally are attributed by the authors to ionic reactions which take place in the N₂ laser plasma.

Thus, the irregularity in the pressure region around 65 torr is attributed to ionic reaction (3), while the irregularity in the pressure region around 110 torr is attributed to ionic reactions (5) and particularly to process III.

The study of ionic reactions and of plasma ionic situation through the laser output energy in general will be studied at length in the future.

6. Acknowledgments.

The authors want to thank warmly Mr. S. Koutsouvelis for his continuous invaluable technical assistance.

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