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#### THEORY OF A CW SUPERSONIC NITROGEN RECOMBINATION LASER

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Abstract.- Quasi-two dimensional model calculations show the possibility to realize a cw atomic nitrogen laser, pumped by the recombination process in rapidly expanded plasma flows. Sufficient gain for lasing is demonstrated for stagnation conditions of 13 000  $\leq$  T  $\leq$  24 000 K and 2  $\leq$  p  $\leq$  30 kPa. Location and size of the gain regimes in the flow direction depend<sup>o</sup>on the plasma starting condition. At a wavelength of 0.91 µm values of small signal gain up to 0.6 m<sup>-1</sup> can be attained. The model is described and some selected results are presented. The effect of the electron heat conduction is discussed.

#### 1. Introduction

The idea to pump a laser by a rapid recombination of electrons into the highly excited electronic states of a neutral or ionic species is rather old.<sup>1</sup> Meanwhile the mechanism of many pulsed lasers has been explained by this process. The suggestion to operate a recombination laser by exploiting the rapid cooling rates, and hence recombination rates, in a supersonic plasma expansion flow has been made nearly as long ago.<sup>2,3</sup> Though basing on a different physical principle, it has always been tempting to regard such a plasmadynamic laser as the natural extension of the powerful infrared gasdynamic lasers into the visible or even ultraviolet wavelength region. Since recombination is a general physical mechanism to preferentially populate high lying electronic levels, the method should be applicable to pump laser transitions in almost any ionizable element or molecule and thus open a wide spectrum of laser wavelengths. Reviews and more detailed discussions may be found in Refs. 4 and 5. Quite a few theoretical models have been developed to describe the population dynamics by the recombination of expanding plasmas, some yielding very promising results. (A more recent compilation may be found in Ref. 6). Also many experiments have been performed and the existence of population inversions has been proven repeatedly. But only Campbell et al.<sup>7</sup> have been able to actually demonstrate 1 ms laser action in an argon plasma, expanded from a pulsed MPD arc source. However, in an attempt by Hoffmann and Hügel<sup>8</sup> to extend these results to cw operation at equal plasma parameters absorption was found in the laser transitions. Today it is not known which steady state processes prevent the recombination laser action and to what extent existing models do not describe the real physics accurately enough.

In this paper a further theoretical model is described and some calculated results are presented for a nitrogen plasma as a potential laser candidate. The model consists of two main parts to compute the recombination and excitation kinetics simultaneously with the gasdynamics along the axis of a two-dimensional plane nozzle flow and is qualitatively described in the next sections. The numerical details will be described elsewhere.<sup>9</sup>

#### 2. Theory

#### 2.1 Gasdynamics

For a maximum recombination rate, an expansion as fast as possible is desired, being inherently more-dimensional. To considerably reduce the computational needs for a complete two-dimensional treatment and yet take account for such a geometry within realistically specified boundary conditions, the following compromise has been adopted.

The sharp corner nozzle is known to produce the fastest nozzle expansion possible. Flow parameters, i.e. effective area ratios and Mach numbers can be infered at any point of the flow field for an isentropic expansion by the method of characteristics. A streamline can then be considered as a one-dimensional streamtube with completely defined boundary conditions. To find the effect of the non-isentropic nature of the flow, brought about by the recombination reaction, the full set of one-dimensional flow equations for the macroscopic state variables is then integrated along the streamtube. As a particular and simple streamtube the flow along the axis of the nozzle is chosen for the present calculations. In the method of characteristics the isentropy of the flow is expressed by the ratio of the specific heats  $\gamma = C_p/C_v$ . After integrating the full equations a variable  $\chi$  can be assigned to any point of

the flow. With this distribution of locally adjusted isentropic coefficients the calculation of the characteristics can be redone to yield a new set of boundary conditions. In general, convergence is reached within less than ten iterations.

Following Appleton and Bray<sup>10</sup> the flow is represented by a three component, quasineutral plasma consisting of neutral particles and of singly-ionized ions and electrons, that can recombine. Maxwellian velocity distributions are assumed. The electrons are allowed to have a temperature different from the heavy particles, but do flow at the same velocity. Hence, the set of conservation equations comprises an equation for the conservation of momentum, for total energy, electron energy, total mass and number density of the electrons no, from which velocity w, heavy particle temperature T, electron temperature Te, number density n are calculated. Further conservation equations for atoms in the various excited states can be added as is considered necessary. In particular, equations for the three ground states of the chosen species nitrogen are provided.

The conservation equations contain terms to account for the dissipation of energy among the different particles, and for the permanent loss of energy from the plasma by radiation. A term for the heat conduction by electrons in axial direction has been introduced in the electron energy equation to check for its relevance. Since this term contains second derivatives, an a priori knowledge of the electron temperature distribution is needed. Again, it is obtained in the successive course of iterations. The calculation is started with a steady state solution of all conservation equations at the point of singularity, i.e. at the throat of the nozzle. Equality of electron- and heavy particle temperature is assumed at this point, but is not mandatory. The calculation is performed up to an arbitrary Mach number, typically of the order of 5 to 6, with the distance from the throat, x, being the independent variable.

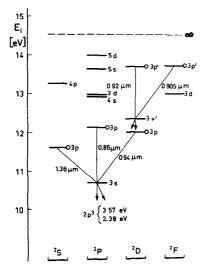
#### 2.2 Excitation kinetics

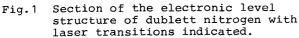
As the element of investigation reported herein atomic nitrogen has been chosen, but other elements like carbon, oxygen etc. have been or may be computed as well. 38 levels for electronic excitation are considered in the model, out of which 24 are treated in separate equations. The kinetic of recombination is treated by the collisonal-radiative decay mechanism, originally introduced by Bates et.al. : The rate equations account for two and three body recombination (ionization) into an arbitrary level i, radiative deexcitation, and transitions induced by electron collisions. Re-absorption of resonance radiation is taken into account by multiplying the spontaneous transition probabilíties with an escape probability factor for the radiation, calculated from the local optical thickness. Probabilities for recombination transitions and transitions due to electron collisions are calculated using formulae given by Drawin for hydrogen<sup>12</sup>, modified with the spectroscopic data for nitrogen. For the

three 2p<sup>3</sup> ground states the transition probabilities are taken from Smith, et.al.<sup>13</sup> The population density n, of every energy level higher than those of the ground states is computed from the algebraic system of the steady state rate equations  $(n_i = 0)$ . Because of the short characteristic times of the levels to acquire the steady state values this approximation proves to be justified everywhere in the expansion, except for the very first steps of computation. This is not the case for the three ground states with energies of  $0. eV ({}^{4}S)$ , 2.38 eV ( ${}^{2}D$ ) and 3.75 eV ( ${}^{2}P$ ). The number densities of these levels n1, n2, n3, together with the number density and temperature of the electrons,  $n_{p}$  and  $T_{p}$ , enter the system of rate equations as the primary, independent variables.

The system can be solved as a separate unit to compute regimes of possible inversions in full generality or, more interestingly, fully coupled to the integration of the flow equations. In the latter case level population and population inversions at any step of integration along the nozzle axis can be infered with regard to location, size and magnitude of the small signal gain.

Fig. 1 shows a section of the Grotrian diagram of the nitrogen dublet with transitions expected to show significant small signal gain at wavelengths ranging from 0.86  $\mu$ m to 1.36  $\mu$ m. In fact, from pulsed experiments these transitions are known as laser lines already.<sup>14</sup> Similar transitions exist in the quartet also. C9-474





#### 3. Numerical results

Calculations were performed for a plane, sharp-edge nozzle with 1 mm throat height. The parameter range for the stagnation temperature and pressure of the starting conditions were 13 333  $\leq$  T<sub>o</sub>  $\leq$  24 000 K and  $2 \leq p_o \leq$  30 kPa. This comprises a degree of ionization from 0.05 to 0.99.

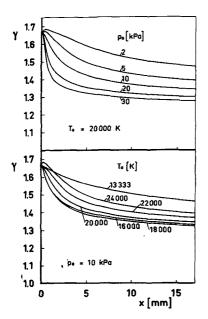


Fig.2 Variation of the quasi-isentropic coefficient  $\gamma$  along the flow direction for various stagnation conditions at the starting point.

Fig.2 demonstrates the non-isentropic nature of the flow, expressed by the variation of the effective isentropic coefficient **y** along the flow direction. The local  $\gamma$  is recalculated from the actual local Mach number and the heavy particle temperature. It is seen that due to the different heat release from the recombination at various starting conditions / drops rapidly to values of 1.3 to 1.5 after a flow distance of 1 cm. Fig. 3 shows, how the recombination of electrons in the strongly non-equilibrium flow tends to freeze the population density of an arbitrary level placed at 12 eV compared to its equilibrium density. Differences in the population or depopulation mechanisms of two levels, connected by an allowed transition can lead to a temporary inversion of the population densities.

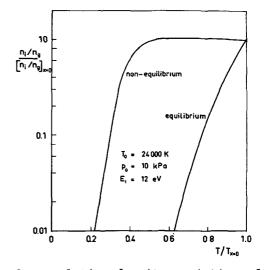


Fig.3 Population density variation of an arbitrary level placed at 12 eV by a slow cooling process (equilibrium) and the rapid cooling process in a non-equilibrium expansion flow.

The calculation of the inversion conditions for possible laser transitions yields parameter ranges of the three dominating variables electron temperature  $T_e$ , electron number density  $n_e$  and ground state number

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density  $n_1$ , that have to be attained simultaneously during the expansion. Fig.4 shows these ranges, valid for the most prominent transitions  $3s'^2D - 3p'^2D^0$  at 918.8 nm and

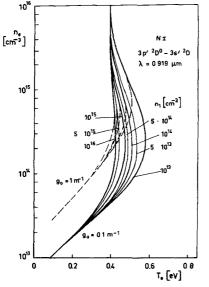


Fig.4 Ranges of small signal gain for the 3p'-3s' transition (steady state ap-proximation for levels i > 1).

 $3s^{2}D - 3p^{2}F^{0}$  at 904.8 nm. The curves indicate parameter triples for which a gain of 0.1  $m^{-1}$  and 1  $m^{-1}$  is to be expected. They reflect the necessity to achieve simultaneously a low electron temperature (below approx. 6000 K) at a sufficiently high electron density (on the order of several  $10^{14}$  cm<sup>-3</sup>), in order to keep the pumping recombination reaction as fast as possible. This requires a close coupling of the electron energy to that of the heavy particles. Depending on the starting conditions, T follows indeed the static flow temperature within a few hundred to fifteen hundred Kelvin. Fig. 5 demonstrates, that for all the starting conditions under consideration the state variables of the electrons change within a rather narrow corridor and do actually assume the states required for population inversion. Representative

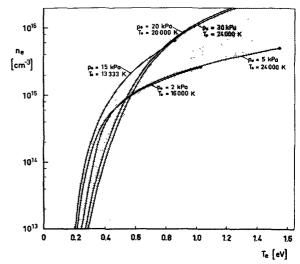


Fig. 5 Change of state of the electrons during expansion.

examples of the ensuing axial gain distribution along the flow direction are displayed in Figs. 6 and 7. Depending on the pressure, the gain tends to appear and attain its maximum rather abruptly and then to fade away as the gas thins out in the continued expansion. In a practical application

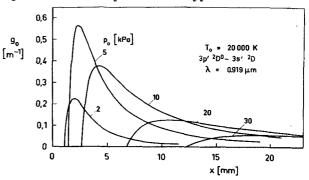


Fig. 6 Small signal gain distribution along the flow direction for various stagnation pressures at the starting point.

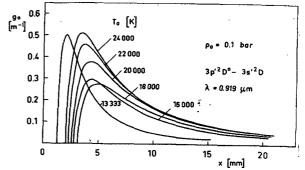


Fig. 7 Small signal gain distribution along the flow direction for various stagnation temperatures at the starting point.

though, a further expansion of the gas seems not appropriate after the maximum of gain is reached. The gain region is shifted downstream as the pressure is raised, while the gain maximum rises with increasing tem-

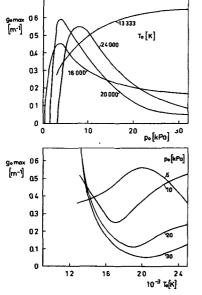


Fig. 8 Variation of maximum small signal gain with the starting conditions.

perature. As recognized from these graphs and also from Fig. 8, where the variation of the gain maximum is plotted as a function of the starting conditions, this rather general behavior is not valid for extremely low starting temperatures. The further and considerable rise of gain at these conditions may be explained from the expansion curves in Fig. 5: the already low starting temperature and the associated small degree of ionization, with the small amount of available ionization energy allows the earlier achievement of a somewhat smaller electron temperature at comparativly higher electron densities, and hence a deeper penetration into the inversion regime occurs.

The maximum small signal gain for this particular transition amounts to approxi-

mately 0.6 m<sup>-1</sup>. As expected, other transitions exhibit gain as well and with the same general behavior. However, the parameter range, for more than 0.1 m<sup>-1</sup> is narrower and the regions are somewhat shifted either up- or downstream. The highest gain is achieved at a distance of 3 to 5 mm downstream of the throat. This corresponds to a flow time of  $5 \cdot 10^{-7}$  s. The other conditions are: velocity w  $\approx 10^4$  m/s, Mach number M $\approx$  3.5, area ratio A/A\* = 6-8, static pressure p  $\approx$ 150 Pa = 1.5 mbar, gas and electron temperature 5300 K and 6000 K, resp.

In order to check for a possible influence of the electron heat conduction on the electron temperature, several test runs have been performed with the inclusion of an appropriate term in the electron energy equation. The result is expressed in Fig.9 as the increase of electron temperature

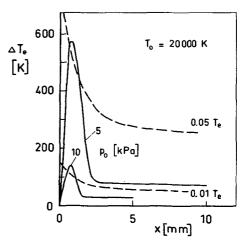


Fig. 9 Local temperature rise of the electrons due to the effect of electron heat conduction along the flow direction.

above the value calculated without this term. It is seen that the effect becomes noticable only within the very first moments of the expansion and has dropped to about 1 % after 2 mm. Since this occurs well before the gain attains its maximum, the effect will be little, if present at all. In fact, because of the reduced recombination rate at the higher electron temperature the electron state distribution along the flow direction remains very nearly unchanged.

#### 4. Experiment

An effort has been made to verify the results in an experiment and to achieve laser oscillation. For this purpose the 5 cathode arc heater, described in Ref. 15 has been operated with pure nitrogen. The results have been negative. In contrast to earlier findings with a plasma of predominantly argon, the flow became extremely inhomogeneous in temperature in the optical direction. Small regions with very hot plasma interchanged with broad regions of cold qas.

#### 5. Conclusions

A computer model has been established to describe the non-equilibrium plasma expansion in a two-dimensional nozzle. Sufficient small signal gain has been found in the computation of a nitrogen model, that should allow the achievement of cw laser oscillation even in a smaller device. An experimental test was not successful, because the plasma source did not produce a flow of sufficient homogenity.

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