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#### IONIC AND THERMAL INSTABILITIES IN E-BEAM PREIONIZED CO<sub>2</sub> EDCL DEVICES IN THE PRESENCE OF A LASER BEAM

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## Ionic and Thermal Instabilities in e-Beam Preionized CO<sub>2</sub> EDCL Devices in the Presence of a Laser Beam (\*).

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Summary. — In the present paper a general model of plasma instabilities in high-power, e-beam-sustained CO<sub>2</sub> laser amplifiers is presented together with a detailed discussion of some experimental cases. This model improves the Haas-Nighan one by allowing explicitly for the various degrees of vibrational freedom and taking into account the laser field. The above model has been used to test the degree of thermal and ionic instability of an EDCL device using four-component gas mixtures (CO<sub>2</sub>-N<sub>2</sub>-He-CO). The stabilizing effect of the laser beam intensity has been assessed, thus finding a dramatic reduction of the growth rate of thermal instabilities.

#### I. - Introduction.

In high-power electric-discharge convection lasers (EDCL) the separation of the plasma ionization from the electron temperature is generally achieved by using a beam of high-energy electrons to create a uniform ionization through-

<sup>(\*)</sup> Work supported by the C.N.R. project on high-power lasers.

out the volume of the plasma. By using the high-energy electrons to provide the ionization, the electron temperature is varied independently by adjusting the electric field (for reviews see ref. (1-3)). If the electron temperature is kept so low that the ionization due to the discharge electrons is small compared to the ionization due to the high-energy electrons of the beam, the discharge remains uniform over the range of pressures investigated. Stable discharge behaviour is obtained when the volumetric electron loss rates due to electronion recombination and electron-neutral attachment equal the volumetric secondary electron production rate.

The electrical power deposited in the plasma is limited by the onset of instabilities which manifest themselves in the form of discharge collapse from a uniform diffuse glow into a constricted arc (see ref. (4-7) for the influence of gas recirculation on the occurrence of plasma instabilities in self-sustained CO. lasers).

The optical output power obtainable from these devices scales with the electrical power input. As the discharge power is increased beyond some levels, laser efficiency falls off rapidly due to gas heating effects and decrease of E/n. Consequently, the discharge specific power cannot be made indiscriminately large. The most prominent instability mode in the externally ionized discharge is the streamer, first observed by Douglas-Hamilton and Mani (8). Moving striations in CO2 laser mixtures were first observed in a self-sustained convection discharge by Nighan et al. (\*) in 1973. Attempts to explain streamers on the basis of electron reproduction rates have been made by many authors (see, e.g., Rogoff (10)) and models for formation and propagation have been proposed (11). Measurements of streamer behaviour in nitrogen discharge have been made by Douglas-Hamilton and Mani (12). Present theories indicate that negative-ion formation in the discharge system and its coupling with the electro A way gases, ionie i. instabi the flo

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<sup>(1)</sup> A. J. DE MARIA: Review of high-power CO2 lasers, in Principles of Laser Plasmas, edited by G. Bekeff (New York, N. Y., 1976).

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<sup>(9)</sup> W. J. Wiegand and W. L. Nighan: Appl. Phys. Lett., 22, 583 (1973).

<sup>(10)</sup> G. ROGOFF: Phys. Fluids, 15, 1931 (1976).

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electron temperature are instrumental in the reduced stability limits (8,13). A way to control the formation of negative ions consists in adding some other gases, such as CO, to the usual components CO2: N2: He. In addition to the ionic instability, HAAS and NIGHAN have shown that the so-called thermal instability mode is always present and can be only contrasted by increasing the flow velocity.

The primary objective of the present paper is to extend the instability analyses of Haas (14) and Nighan (15-18) by accounting for the different vibrational modes of a CO2-N2-He-CO gas laser mixture, while the secondary objective is to calculate and discuss the growth times of the most dangerous instability modes that likely occur in some typical e-beam-sustained  $\mathrm{CO}_2$  lasers. Particular attention is paid to the stabilizing effect of the laser beam. A plasma model similar to that developed by HAAS (14), and simplified by NIGHAN (15), has been adopted. The plasma parameters (secondary ionization and attachment coefficients, etc.) have been calculated by using a four-component Boltzmann numerical code, which has been tested by a Monte Carlo simulation of the discharge.

In contrast to HAAS (14), the model discussed in the following allows for the various vibrational modes. By considering a system whose only components are atoms and diatomic molecules of a single type, the thermal instabilities discussed by HAAS and NIGHAN depend on one vibrational temperature only. In the CO2 system, however, there are at least two vibrational temperatures. In the present model both these temperatures have been considered. Starting from the fluid dynamic and kinetic equations and making use of the linear perturbation theory, we have obtained a system of equations governing the temporal and spatial evolution of the perturbation amplitudes of the electronic density and the vibrational and electronic temperatures. This set of equations has been used to eliminate the perturbation amplitudes and shown to result in a fifth-order equation for the growth rates of thermal and ionic instabilities. The coefficients of the equation are expressed in terms of the (theoretically) predicted steady-state plasma properties.

The stabilizing effect of the laser beam has been studied introducing a radiation beam of constant intensity. The laser beam depopulates by stimulated emission the upper laser level, thus providing an additional channel for the disposal of the vibrational energy stored in the CO2 asymmetric stretching mode without heating the gas.

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The organization of the present paper is as follows. In sect. 2, the particle and photon kinetics in gas lasers is discussed. The laser system model is illustrated in sect. 3. Fluid dynamics and molecular kinetics of a four-component CO<sub>2</sub> laser are discussed in sect. 4. A linearized stability analysis is presented in sect. 5. Section 6 contains a detailed account of the numerical determination of the plasma parameters which control laser stability. In the same section the results of some numerical calculations are also presented and discussed.

#### 2. - Particle and photon kinetics in gas lasers.

In EDL-type CO<sub>2</sub> lasers a glow discharge is the mechanism responsible for heating the gas mixture and, eventually, generating electrons. These undergo collisions with molecules of the laser mixture and thereby convert some of their energy into molecule translation, rotation, vibration and electron-binding energy. A macroscopic description of these processes requires averaging each type of collision over the velocity distribution both of electrons and heavy particles. The velocities of these components undergo continuous redistribution, subject to ensuring on the average a) zero overall charge, b) stationary total energy, c) conservation of the mean number of particles. A slight departure from the equilibrium configuration can be described by a set of rate equations.

2.1. Vibrational excitation by electron collisions. – The electron-M<sub>2</sub> collision process is described by a reaction of the type

(1) 
$$e + M_2(v) \xrightarrow{k_c^v} e + M_2(v+1)$$
,

where  $M_2(v)$  denotes a diatomic molecule (i.e.  $N_2$ , CO,  $O_2$ , NO) having v quanta of vibrational energy. As a result of the collision, the molecule jumps to the next higher energy level.

In  $O_2$ ,  $N_2$ , CO and NO molecules, vibrational excitation occurs through the formation of a negative ion, fairly stable as a result of shape resonance (see ref. (19)). Triatomic molecules, such as  $CO_2$ , also undergo a similar process. The lifetime and energy of these resonances affect the behaviour of the cross-sections and rate constants of these excitation processes.

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<sup>(19)</sup> G. J. Schulz: A review of vibrational excitation of molecules by electron impact at low energies, in Principles of Luser Plasmas, edited by G. Bekefi (New York, N. Y., 1976).

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impact at Y., 1976). 2.2. Electron energy distribution. – Reaction (1) is the main channel through which the laser is pumped: the electrons, accelerated by the E field, impart some of their electric energy JE to the gas mixture by exciting  $N_2$  (v=0) molecules to the v=1 level. Assessing the fraction of power that is converted to vibrational energy requires the knowledge of the electron velocity distribution  $f_{\bullet}(v_{\bullet})$ . If we assume single scattering, that the mutual repulsion between electrons is negligible (low electron density) and that the distribution laws  $f_{\rm m}$  of the molecules over the various vibrational levels are known, we may calculate  $f_{\bullet}$  by solving a Boltzmann equation in which the cross-sections for elastic collisions  $(Q_{\rm m})$ , involving only momentum transfer, and the cross-sections for inelastic collisions (vibrational excitation  $Q_{\rm v}$ , ionization  $Q_{\rm i}$ , attachment  $Q_{\rm s}$ ) occur as known functions of  $u_{\rm e}$ .

Matters are considerably simplified if the distributions  $f_{\rm m}$  for atoms and molecules are assumed to be Maxwellian, at temperature T. Once  $f_{\rm e}$  is known, the rate constants  $k_{\rm e}^{\rm v}$ ,  $k_{\rm e}^{\rm i}$  and  $k_{\rm e}^{\rm s}$  corresponding to the forward processes may be calculated by numerical means. In the case of electron collisions of relatively high energy, to a good approximation collisions of the second kind, in which energy is transferred to the electrons by a heavy particle, may be neglected. Such an approximation is tantamount to setting the backward rate constant  $k_{\rm b}$  equal to zero in eq. (1).

 $2^{\circ}3$ . Molecular collisions. – Vibrationally excited  $N_2$  molecules transfer their energy to  $CO_2$ , which is brought to the  $00^{\circ}1$  level. From this point, the controlling role in the process is no longer held by electron-molecule collisions, but it is taken over by molecule-molecule collisions. The interaction between  $N_2$  and  $CO_2$  is described by the equation

(2) 
$$N_2(v=1) + CO_2(00^00) \xrightarrow{k_t} N_2(v=0) + CO_2(00^01)$$
.

In this case, both directions of the reaction are possible and the rates  $k_i$  and  $k_b$  are related to one another by Arrhenius law  $k_b/k_i = \exp{[-E_b/kT]}$ , where the activation energy  $E_a$  equals  $-14~\mathrm{cm}^{-1}$ , k is the Boltzmann constant and T is the temperature of the reaction partners.

The energy transferred to CO<sub>2</sub> tends to be redistributed, as a result of collisions, among the various levels (V-V transfer) and to be converted to traslational and rotational energy (V-T transfer). The populations of the various levels may be assumed to be described by Boltzmann distributions, each vibrational degree of freedom of the laser mixture being associated with its own temperature. As a result, the Boltzmann equation for molecules reduces to a system of rate equations for the traslational and vibrational temperatures. In particular, this system will contain terms connected with photon-molecule

collisions, which govern the origin and evolution of an electromagnetic field within the laser cavity. This photon collision model for the interaction with radiation implies the assumption that coherence effects are absent and radiation is isotropic. The former of these assumptions cannot be removed by any ad hoc correction, whereas the latter can be corrected by dividing the Einstein coefficients by  $4\pi$ .

2.4. Rate equations and stability. - The analysis of an electric-discharge laser reduces to solving a set of Boltzmann equations for electrons, photons and the various vibrational degrees of freedom, respectively. The form of these distribution functions is decided by the vibrational temperatures and the corresponding Boltzmann equations reduce to rate equations. In particular, the Boltzmann equation for the laser field photons, because of the near monochromatic conditions, reduces to the I(r) radiative-transport equation within the optical eavity, filled by a medium whose gain depends on the same I(r) and the saturation intensity I.

Once the set of kinetic equations has been solved, it will be possible to calculate for a given discharge volume geometry the beam power as a function of the electric power delivered and the thermo-fluid-dynamic parameters. To complete the picture, the stability of the system as a whole must be analysed. In fact, since not all of the quantities which influence the output power lend themselves to being controlled, their possible fluctuations need to be taken into account. Should a regenerative process initiate, the state of the system is likely to undergo profound modifications, including those which will cause lasing to cease. In the worst cases, the glow discharge may collapse and go over to an arc discharge.

A typical case of instability arises when the laser is switched on. As long as the electric power remains below a certain threshold, the glow discharge will be stable even though without photons. On exceeding the threshold, the glow discharge becomes unstable, the system evolving towards a different state in which photons are also present. The population inversion builds up as the electron temperature increases. Slight field strength fluctuations, due to spontaneous photon emission by excited molecules, initiate an amplification process that alters the composition of the initial system (it adds a high photon concentration to the electrons and heavy particles, the only components initially present) and modifies the populations of the various levels. If care is taken to let the electric power grow slowly enough for the system to evolve through successive states of equilibrium, the beam power will be found to increase steadily. Eventually a second threshold is exceeded as evidenced by large intensity fluctuations, an indication that the system is approaching an unstable configuration beyond which the laser source becomes unable to perform regularly and efficiently.

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#### 3. - Laser system model.

The laser medium response to the variation of one of the local state parameters may give rise to a regenerative process involving a significant part of the same medium and leading to a permanent alteration of the system. Similar effects occur when the system is found under unstable equilibrium conditions. In a continuous medium the classification of the various types of instabilities which can arise requires more attention than is required for systems having a discrete number of degrees of freedom (see ref. (20-24)). The role of convective effects has been extensively discussed by STURROCK (see ref. (21)). The velocity with which a perturbation propagates may, in fact, be found to be smaller than the medium recycling velocity; in this case the perturbation is eliminated by convection and its negative effect on the overall system stability is neutralized. This mechanism lends itself to be used as a means for dynamically stabilizing an intrinsically unstable system. It is well known that, in any case, by whatever means the state parameters are controlled, the laser mixture will always exhibit some measure of thermal instability. To prevent the glow discharge from collapsing, we can only act on the recycling flow velocity and thereby ensure that the residence time in the cavity is smaller than the perturbation build-up time.

Among the various parameters that define the state of the system, the beam power is the most significant. Thus, in characterizing the system equilibrium, possible beam power fluctuations should be taken into account, also in view of the fact that their magnitude is correlated with the distance from the nearest nonequilibrium configuration. How matters stand in this respect is well exemplified by what occurs in a common phase transition.

Turbulence in a fluid is fairly well characterized by the spectrum of its density, pressure or velocity fluctuations. The case of radiation emitted by a lasing medium is not dissimilar as, generally speaking, its spectrum is very narrow and it is this narrowness which accounts for many of the merits of laser devices. While turbulence associated with density fluctuations is responsible for thermo-fluid-dynamic dissipative effects, turbulence of the cavity field promotes power redistribution over several transverse modes, which detracts from transverse coherence of the wave front of the radiation.

When the laser system approaches a configuration corresponding to the transition to a different macroscopic state, the spectrum of the radiation emitted undergoes profound alterations. Near the laser oscillation threshold,

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<sup>(20)</sup> C. NORMAND, Y. POMEAU and M. G. VELARDE: Rev. Mod. Phys., 49, 581 (1977).

<sup>(21)</sup> F. Cap: Handbook of Plasma Instabilities (New York, N. Y., 1978).

<sup>(22)</sup> P. GLANDSDORFF and I. PRIGOGINE: Thermodynamic Theory of Structure, Stability and Fluctuations (London, 1971).

<sup>(23)</sup> Instability of continuous systems, in Iutam Symposium Herrenale, 1969, edited by H. Leiphalz (Berlin, 1971).

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the spectrum is still bread enough to prevent resolving any particular spectral structure. Above threshold, there will be a line spectrum, denoting the presence of several modes which escillate independently of one another and contribute incoherently to the formation of the overall spectrum. This is further evidence for instability, the impossibility to lock the various modes with one another, so that we must resign ourselves to having the power distributed over several modes, to the detriment of the peak power obtainable. Often, in addition to fluctuations due to different modes beating with one another, mechanical vibrations contribute to further increase fluctuations by making the opticalcavity length to vary with time. A distinct feature of the latter spectrumbroadening mechanism is the fact that it need not necessarily be associated to any instability, in the sense which is commonly attributed to this term. An analogous situation occurs with recycling-flow lasers where, under certain conditions, the beam power is modulated at a frequency which is a multiple of the reciprocal of the laser channel transit time. In these lasers, such a fluctuation is also a comparatively steady feature, though its magnitude may occasionally become so large to convert the c.w. to a pulsed laser. This behaviour of recycling-flow lasers is due to competition between a) the mixture regeneration processes (with supply of fresh lasing material) and b) relaxation processes due to the same laser field and to collisions processes involving active molecules. A somewhat similar picture occurs with relaxation oscillations engendered by gain inhomogeneity.

All of the above instability mechanisms involve the cavity field and, in accord with the general classification scheme suggested by Lehnert (see ref. (21)), may be classified as photon-medium-type instabilities.

Lasers may also present other instabilities, due to gas-dynamical or plasmachemical factors. In these cases, the extent of unbalance will again be apparent as beam power fluctuations, even though in this respect the role of the field is merely passive. A detailed analysis of the various instabilities of this type, that may occur in electrically excited molecular flow lasers (with either self-sustaining or electron-beam-assisted discharges), was first given by HAAS and then improved by Nighan. Allowing for the characteristic parameters of these systems, they succeeded in classifying the instabilities in terms of characteristic frequencies, build-up times and onset mechanisms. NIGHAN showed, in particular, that ionic-type instabilities (due to negative-ion generation and neutralization processes in the discharge) and thermal instabilities (due to local increase of the translational temperature) are the main mechanisms responsible for the origin of striations and filaments and for glow discharge collapse, respectively. All these effects conspire in setting a limit to the maximum density of the electric power delivered to the mixture with the consequent limitation of the obtainable optical power.

Whereas ionic-type instabilities affect the beam only slightly, thermal-type instabilities involve the whole system and make the beam to follow the same

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A way proach and It should a to energy t degrees of 1 of the detrilines of the changes of the other parameters. Negative-ion attachment instability is apparent in a certain hardness of the discharge, which will exhibit striations; by contrast, thermal-type instability develops almost exponentially with time, leading to a fast collapse of the glow discharge, which would turn into an arc discharge were it not for gas-recycling flow which cuts its diffusion, thus enabling the laser to work.

The latter instability does affect the radiation emitted, whose spectrum is found to be altered. The study of this effect is accordingly important for the purposes of a more correct analysis of the laser radiation spectrum. Specific questions are how does the saturation intensity, the line width, the beam power, etc. vary as a function of the electric power supplied to the laser. Unfortunately, these questions can only be answered in qualitative terms. The only possible theoretical predictions have to do with the likely increase or decrease of these quantities for given variations of the electric power and of other parameters.

As already mentioned in the introduction, it is the purpose of the present paper to analyse a laser system model containing a number large enough of parameters of state to account for the main types of instability. In particular, we have striven to account for the laser beam coupling with the recycling flow and for the consequences of the ionic and thermal instabilities investigated by HAAS and NIGHAN. In contrast to HAAS, the present model allows for the various degrees of vibrational freedom explicitly, two distinct aims being envisioned. In the first place, by considering a system whose only components are atoms and diatomic molecules of a single type, in the Haas-Nighan model there is no difficulty in defining a vibrational temperature. In the CO<sub>2</sub> system, however, there are at least two vibrational temperatures, each exceeding the traslational temperature. That corresponding to the 10% lower laser level lies between 400 and 600 K, whereas that of the upper level may reach 5000 K.

A  $CO_2(00^\circ 1)$  molecule transfers its energy first to the lower level 100 via the almost resonant VV reaction  $CO_2(00^\circ 1) \to CO_2(110)$ . Then  $CO_2(110)$  loses its vibrational energy by a VT collision with a partner M, mostly He. This two-step process is characterized by two rate constants, namely  $k_{vv}$  and  $k_{vr}$ , which have a different dependence on the translational temperature T. Consequently, when this complex process is reduced to an equivalent VT relaxation, the values of the relaxation and its temperature derivative are not clearly defined.

A way out of the deadlock should be sought by tracing back Haas' approach and trying to remove as many as possible of the simplifying assumptions. It should also be mentioned that Haas-Nighan model considers no effects due to energy transfer, whereas it cannot be ruled out that communication among degrees of freedom at different temperatures might contribute to reduce some of the detrimental effects predicted by Haas. At the same time, the general lines of the present analysis have been kept fairly close to those of Haas-Nighan,

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#### 4. - Theoretical analysis.

As already mentioned, the present analysis is aimed to extend the Haas-Nighan model of discharge instabilities in  $\mathrm{CO_2}$  EDCL devices. It relies on a set of equations which can be conveniently separated into three major parts: fluid dynamics, molecular kinetics and discharge characteristics (see, e.g., (25)). The attention will be focused on the second part, where the complex evolution of the  $\mathrm{CO_2\text{-}N_2\text{-}CO\text{-}He}$  system comes into play, thus making the departure from the above model more striking.

41. Fluid dynamics. - Due to the different roles played by negative and positive ions, a set of density equations for the main electrically active species is used:

(3) 
$$\frac{\mathrm{D}}{\mathrm{D}t}n + n\nabla \cdot \boldsymbol{u} = 0,$$

(4) 
$$\frac{\mathrm{D}}{\mathrm{D}t} n_{\mathrm{p}} + n_{\mathrm{p}} \nabla \cdot \boldsymbol{u} + \nabla \cdot (n_{\mathrm{p}} \boldsymbol{u}_{\mathrm{p}}) = n_{\mathrm{e}} n k_{\mathrm{i}} - n_{\mathrm{e}} n_{\mathrm{p}} k_{\mathrm{r}}^{\mathrm{e}} - n_{\mathrm{p}} n_{\mathrm{n}} k_{\mathrm{r}}^{\mathrm{i}} + S_{\mathrm{ext}}^{\mathrm{ext}},$$

(5) 
$$\frac{\mathrm{D}}{\mathrm{D}t} n_{\mathrm{n}} + n_{\mathrm{n}} \nabla \cdot \mathbf{u} + \nabla \cdot (n_{\mathrm{n}} \mathbf{u}_{\mathrm{n}}) = n_{\mathrm{o}} n k_{\mathrm{a}} - n_{\mathrm{p}} n_{\mathrm{n}} k_{\mathrm{r}}^{\mathrm{i}} - n_{\mathrm{n}} n k_{\mathrm{d}},$$

(6) 
$$\frac{\mathrm{D}}{\mathrm{D}l} n_{\mathrm{e}} + n_{\mathrm{e}} \nabla \cdot \mathbf{u} + \nabla \cdot (n_{\mathrm{e}} \mathbf{u}_{\mathrm{e}}) = \\ = n_{\mathrm{e}} n k_{\mathrm{i}} - n_{\mathrm{e}} n_{\mathrm{p}} k_{\mathrm{r}}^{\mathrm{e}} - n_{\mathrm{u}} n k_{\mathrm{a}_{1}} - n_{\mathrm{e}} n^{2} k_{\mathrm{a}_{2}} + n_{\mathrm{n}} n k_{\mathrm{d}} + S_{\mathrm{ext}},$$

where  $D/Dt = \hat{c}/\hat{c}t + u \cdot \nabla$ , u is the mass average velocity,  $u_n$ ,  $u_p$  and  $u_e$  are the diffusion velocities of the ionic species and electrons. The rate coefficients k represent the main ion production and loss processes: electron impact ionization of molecules  $(k_i)$ , electron-molecules attachment  $(k_a)$ , two-body electron-ion recombination  $(k_r^e)$ , positive-ion-negative-ion recombination  $(k_r^e)$  and detachment by neutral impact  $(k_d)$ . Ionization of electronically excited species has been omitted for simplicity. An independently controlled source of ionization and excitation in the form of an e-beam has been designated by  $S_{ext}$ . For the operating conditions of typical e-beam lasers,  $S_{ext}$  is given by

$$S_{\rm ext} = 3.92 \cdot 10^{16} \, pj_{\rm EB} E_{\rm EB}^{-0.7} (6.9 X_{\rm CO_2} + 3.7 X_{\rm N_2} + 4 X_{\rm CO} + 0.65 X_{\rm He}) \, \rm cm^{-3} \, s^{-1} \, ,$$

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<sup>(25)</sup> M. J. YODER, H. H. LEGNER, J. H. JACOB and D. R. HOUSE: J. Appl. Phys., 49, 3171 (1978).

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e) cm<sup>-3</sup> s<sup>-1</sup>,

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 $E_{\rm EB}$  being the e-beam energy in keV,  $j_{\rm EB}$  the relative current density in mA/cm<sup>2</sup> and p the gas pressure in Torr.  $X_{\rm A}$  will indicate hereinafter the concentration of the molecular component A.

Due to the fast electron relaxation processes, the plasma is assumed to be neutral (14) on the time scale of the instabilities discussed in this paper. This, in turn, implies  $\nabla \cdot E = 0$ .

The momentum equation is expressed by

(8) 
$$\varrho \frac{\mathrm{D}}{\mathrm{D}t} \boldsymbol{u} = -\nabla p + \mu \nabla^2 \boldsymbol{u} + \left(\mu_{\mathrm{B}} + \frac{1}{3}\mu\right) \nabla(\nabla \cdot \boldsymbol{u}) + \varrho_{\mathrm{c}} \boldsymbol{E},$$

where  $\varrho$  is the mass density of the mixture. The contribution of the force acting on the space charge (cf. ref. (14), eq. (5)) has been neglected.  $\mu$  and  $\mu_{\rm B}$  are the kinematic and bulk viscosity coefficients, respectively.  $\mu$  can be approximated by the Sutherland equation, which for the mixtures of interest reads (see ref. (26))  $\mu = 1.44 \cdot 10^{-5} T^{\frac{3}{2}}/(111 + T)$ .

42. Molecular kinetics. – The CO<sub>2</sub> laser kinetics has been discussed by many authors. The Landau-Teller equations have been used for describing the energy stored in the vibrational modes of CO<sub>2</sub>, N<sub>2</sub> and CO. Implicit in these equations is the assumption of the partial thermal equilibrium for the rotational and vibrational degrees of freedom. Essential to the mechanism of energy relaxation and exchange is the photon deactivation of the upper laser level to the lower one. When no laser power is extracted, the primary loss mechanism is the collisional deactivation of CO<sub>2</sub>(001) by helium, N<sub>2</sub> and CO to the (030) level. The energy transfer to and from the coupled degrees of freedom is described by the following set of equations:

(9) 
$$\frac{\mathrm{D}}{\mathrm{D}t}(nE_{\mathrm{TR}}) + (nE_{\mathrm{TR}} + p)\,\nabla\cdot\boldsymbol{u} = \varkappa\nabla^2T + \mu\varphi + n_{\bullet}n(\nu_{\mathrm{m}}/n)\,kT_{\bullet} + n\dot{E}_{\mathrm{TR}(\mathrm{VT})}\,,$$

(10) 
$$\frac{\mathbf{D}}{\mathbf{D}t} (n_{\alpha} E_{\mathbf{v}}^{\alpha}) + n_{\alpha} E_{\mathbf{v}}^{\alpha} \nabla \cdot \mathbf{u} =$$

$$= k_{\mathbf{v}}^{\alpha} \nabla^{2} T_{\alpha} + n_{\epsilon} n(v_{\mathbf{v}}^{\alpha}/n) k T_{\epsilon} - n_{\alpha} \dot{E}_{\mathbf{v}(\mathbf{v}\mathbf{r})}^{\alpha} - n_{\alpha} \dot{E}_{\mathbf{v}\mathbf{v}\mathbf{r}}^{\alpha} - n_{\alpha} \dot{E}_{\mathbf{v}\mathbf{v}$$

In the translation-rotation energy equation  $E_{\rm TR} = (\frac{5}{2} - X_{\rm Re})kT$ ,  $\varkappa$  is the thermal conductivity,  $\varphi$  the viscous dissipation function and k the Boltzmann constant.  $r_{\rm m}({\rm s}^{-1})$  represents the translation-rotation contribution to the total electron energy exchange collision frequency  $r_{\rm u}$ .  $\vec{E}_{\rm TR(VT)}$  represents the rate of energy change by collisional deactivation. Following the kinetic model worked

<sup>(26)</sup> J. O. Hirschfelder, C. F. Curtiss and R. B. Bird: Molecular Theory of Gases and Liquids (New York, N. Y., 1954).

out originally by Gordietz et al. (27) in 1968 and improved since then by many authors in order to fit the special features displayed by different CO<sub>2</sub> laser devices, we put

(11) 
$$\dot{E}_{\text{TR}(\text{VT})} = X_{\text{CO}_{\bullet}} \left( \frac{E_{\text{V}}^{1}(T_{1}) - E_{\text{V}}^{1}(T)}{\tau_{10}(T)} + \frac{E_{\text{V}}^{2}(T_{2}) - E_{\text{V}}^{2}(T)}{\tau_{20}(T)} \right) = \dot{E}_{\text{V}(\text{VT})}^{1} ,$$

where

$$(12) \tau_{20}(T) = n^{-1} \{ X_{\text{CO}_2} K_{20}^{\text{CO}_2}(T) + X_{\text{N}_2} K_{20}^{\text{N}_2}(T) + X_{\text{CO}} K_{20}^{\text{CO}}(T) + X_{\text{He}} K_{20}^{\text{He}}(T) \}^{-1}$$

is the relaxation time of the bending mode and, according to MANES and SEGUIN (28),  $\tau_{10} \simeq 4.5\,\tau_{20}$  is the relaxation time of the symmetric stretching mode.

In the vibrational energy equation  $E^{\alpha}_{\mathbf{v}}(T_{\alpha})$  is the average energy

(13) 
$$E_{\mathbf{v}}^{\alpha}(T_{\alpha}) = \varepsilon_{\alpha} Q_{\mathbf{v}}^{\alpha}(T_{\alpha}) \exp\left[-\varepsilon_{\alpha}/kT_{\alpha}\right]$$

of the CO<sub>2</sub> symmetric stretching ( $\alpha=1$ ), bending ( $\alpha=2$ ), asymmetric ( $\alpha=3$ ), and N<sub>2</sub> ( $\alpha=4$ ) and CO( $\alpha=5$ ) modes.  $\varepsilon_{\alpha}$  is the quantum of vibrational energy and  $Q_{\rm v}^{\chi}(T_{\alpha})$  the  $\alpha$ -mode partition function. In accordance with the present labelling  $n_1=n_2=n_3=n_{\rm CO_1},\, n_4=n_{\rm N_2}$  and  $n_5=n_{\rm CO}$ .  $T_{\alpha}$  represents the vibrational temperature and  $k_{\rm v}^{\alpha}$  the conductivity of the  $\alpha$ -mode vibrational energy.  $r_{\rm v}^{\chi}$  indicates the contribution to the total electron energy exchange collision frequency  $r_{\rm u}$ . With  $E_{\rm v(vT)}^{\alpha}$ ,  $E_{\rm v(vv)}^{\alpha}$  and  $E_{\rm v(ph)}^{\alpha}$  we represent the rate of change of the  $\alpha$ -mode vibrational energy by collisional relaxation (VT), energy exchange (VV) and photon emission or absorption. These quantities replace the factor  $\{E_{\rm v}(T_{\rm v})-E_{\rm v}(T)\}/\tau_{\rm vT}$  describing the molecular kinetics in the Haas paper (cf. eqs. (6) and (8) in ref. (14)). They are here represented by simple expressions, that is

$$\begin{cases} \dot{E}_{\text{V(VT)}}^{1} \equiv \frac{JE}{n_{\text{CO}_{2}}} F_{\text{VT}}^{1} = \frac{E_{\text{v}}^{1}(T_{1}) - E_{\text{v}}^{1}(T)}{\tau_{10}} , \\ \dot{E}_{\text{V(VT)}}^{2} \equiv \frac{JE}{n_{\text{CO}_{2}}} F_{\text{VT}}^{2} = \frac{E_{\text{v}}^{2}(T_{2}) - E_{\text{v}}^{2}(T)}{\tau_{20}} , \\ \dot{E}_{\text{V(VT)}}^{3} \equiv \dot{E}_{\text{V(VT)}}^{4} \equiv \dot{E}_{\text{V(VT)}}^{5} = \dot{E}_{\text{v}}^{5}(\tau_{1}) = 0 , \\ \dot{E}_{\text{v}(\text{vv})}^{1} \equiv \frac{JE}{n_{\text{CO}_{2}}} F_{\text{vv}}^{1} = -\frac{E_{\text{v}}^{1}(T_{1}) - E_{\text{v}}^{1}(T_{2})}{\tau_{12}} + \frac{\nu_{1}}{\nu_{3}} \frac{E_{\text{v}}^{3}(T_{3}) - E_{\text{v}(\text{eq})}^{3}}{\tau_{3}} , \\ \dot{E}_{\text{v}(\text{vv})}^{2} \equiv \frac{JE}{n_{\text{CO}_{1}}} F_{\text{vv}}^{2} = \frac{E_{\text{v}}^{1}(T_{1}) - E_{\text{v}}^{1}(T_{2})}{\tau_{12}} + \frac{\nu_{1}}{\nu_{3}} \frac{E_{\text{v}}^{3}(T_{3}) - E_{\text{v}(\text{eq})}^{3}}{\tau_{3}} , \end{cases}$$

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<sup>(27)</sup> B. F. GORDIETZ, N. N. SOBOLEV, V. V. SOKONIKOV and L. A. SHELEPIN: IEEE J. Quantum Electron., QE-4, 796 (1968).

<sup>(28)</sup> K. R. MANES and H. J. SEGUIN: J. Appl. Phys., 43, 5073 (1972).

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ic ( $\alpha = 3$ ), nal energy is present the vibraal energy, e collision change of nergy exeplace the Haas by simple IONIC AND THERMAL INSTABILITIES ETC.

$$\begin{split} E_{\mathbf{v}(\mathbf{v}\mathbf{v})}^{3} &\equiv -\frac{JE}{n_{\mathbf{co}_{1}}} F_{\mathbf{v}\mathbf{v}}^{3} = -\frac{E_{\mathbf{v}}^{3}(T_{3}) - E_{\mathbf{v}(\mathbf{eq})}^{3}}{\tau_{3}} + \\ &\qquad \qquad + \frac{E_{\mathbf{v}}^{4}(T_{4}) - E_{\mathbf{v}}^{4}(T_{3})}{\tau_{43}} + \frac{E_{\mathbf{v}}^{5}(T_{5}) - E_{\mathbf{v}}^{5}(T_{3})}{\tau_{53}}, \\ E_{\mathbf{v}(\mathbf{v}\mathbf{v})}^{4} &\equiv \frac{JE}{n_{\mathbf{co}_{4}}} F_{\mathbf{v}\mathbf{v}}^{4} = -\frac{E_{\mathbf{v}}^{4}(T_{4}) - E_{\mathbf{v}}^{4}(T_{3})}{\tau_{43}} - \frac{E_{\mathbf{v}}^{4}(T_{4}) - E_{\mathbf{v}}^{4}(T_{5})}{\tau_{45}}, \\ E_{\mathbf{v}(\mathbf{v}\mathbf{v})}^{5} &\equiv -\frac{JE}{n_{\mathbf{co}_{4}}} F_{\mathbf{v}\mathbf{v}}^{5} = -\frac{E_{\mathbf{v}}^{5}(T_{5}) - E_{\mathbf{v}}^{5}(T_{3})}{\tau_{53}} + \frac{E_{\mathbf{v}}^{4}(T_{4}) - E_{\mathbf{v}}^{4}(T_{5})}{\tau_{45}}, \end{split}$$

where JE is the electric power deposited per unit volume. The quantities  $F_{\text{VT}}^{\alpha}$ ,  $F_{\text{VV}}^{\alpha}$  have been defined in such a way as to result positive under normal operating conditions of the laser. The energy  $E_{\text{V(eq)}}^3$  stands for  $h\nu_3\{\exp\left[\theta_1+\theta_2+\Delta E/kT\right]-1\}^{-1}$  with  $\Delta E=h(\nu_3-\nu_2-\nu_1)=-423$  K, h is the Planck constant,  $\nu_{\alpha}$  the frequencies of the three vibrational modes of the CO<sub>2</sub> considered,  $\theta_{\alpha}=\varepsilon_{\alpha}/kT_{\alpha}$  and  $\tau_3^{-1}=n_{\text{CO}_4}A_3(T)\{\exp\left[-423/T\right]-\exp\left[-\theta_1-\theta_2\right]\}$  (as regards the coefficient  $A_3(T)$ , see appendix B). For the laser field contribution we have

(15) 
$$\dot{E}_{V(ph)}^{1} = -\frac{\nu_{1}}{\nu_{3}} \dot{E}_{V(ph)}^{3} = -h\nu_{1} \frac{\Delta N}{n_{CO_{3}}} WI = -\frac{JE}{n_{CO_{3}}} F_{phV}^{1},$$

the other terms being identically zero. W is the stimulated emission rate on line centre, which is proportional to the pressure through the line width factor.  $\Delta N$  represents the population inversion, which for a simple line laser, is given by

(16) 
$$\Delta N = \frac{n_{\text{co}_1}}{Q_1 Q_2 Q_3} \left\{ P(J) \exp \left[ -\theta_3 \right] - P(J+1) \frac{\theta_J}{\theta_{J+1}} \exp \left[ -\theta_1 \right] \right\},$$

where  $P(J) = (2hcB/kT)\Theta_J \exp\left[-hcBJ(J+1)/kT\right]$ ,  $\Theta_J = 2J+1$ ,  $J = \text{rotational quantum number and } B = 0.4 \text{ cm}^{-1} = \text{rotational constant.}$  I is the laser field intensity and c the velocity of light.

For typical operating conditions we can group the five modes in two sets, labelled 1 and 3. The parameters F and  $\tau$  relative to these two groups will be indicated by adding a tilde in order to avoid confusion with the initial ones.

#### 5. - Stability analysis.

The search for instability modes can be based on the linear stability theory at least for assessing the role of the plasma parameters and laser intensity in controlling the threshold and the initial growth rate. As usual we expand the parameters in the form  $\psi(1+\psi')$ , leading to first-order equations for the

perturbations of the electron and negative-ion densities, of the traslational, vibrational and electron temperature. Then, Fourier-transforming these equations, after some algebra (an  $\exp\left[i(\omega t - k \cdot r)\right]$  time and space dependence is assumed), we obtain the system

(17) 
$$r_{k}(n_{e^{\pm}} + T_{k}) = -\left(\frac{n_{n}}{n_{e}} \frac{1}{\tau_{d}} + \frac{n_{e}}{n_{p}} \frac{1}{\tau_{r}^{e}} \div \frac{S}{n_{e}}\right) n_{e^{\pm}} + \left(\frac{n_{n}}{n_{e}} \frac{1}{\tau_{d}} - \frac{n_{n}}{n_{p}} \frac{1}{\tau_{r}^{e}}\right) n_{n^{\pm}} - \left(\frac{1}{\tau_{r}^{e}} + \frac{n_{n}}{n_{e}} \frac{\tau_{d,T}}{\tau_{d}} - \frac{1}{\tau_{a_{2}}}\right) T_{k} + \left(-\frac{\tau_{i,\tau_{e}}}{\tau_{i}} + \frac{\tau_{i,\tau_{e}}}{\tau_{r}^{e}} + \frac{\tau_{a_{1},\tau_{e}}}{\tau_{a_{1}}} + \frac{\tau_{a_{1},\tau_{e}}}{\tau_{a_{2}}}\right) T_{e^{\pm}} ,$$

$$(18) r_k(n_{nk} + T_k) = \left\{ \frac{n_e}{n_n} \left( \frac{1}{\tau_{a_1}} + \frac{1}{\tau_{a_2}} \right) - \frac{n_e}{n_p} \frac{1}{\tau_r^i} \right\} n_{ek} - \left\{ \frac{n_n}{n_p} \frac{1}{\tau_r^i} + \frac{n_e}{n_n} \left( \frac{1}{\tau_{a_1}} + \frac{1}{\tau_{a_2}} \right) \right\} n_{nk} - \frac{n_e}{n_n} \left( \frac{\tau_{a_1, \tau_e}}{\tau_{a_1}} + \frac{\tau_{a_2, \tau_e}}{\tau_{a_2}} \right) T_{ek} - \left\{ \frac{1}{\tau_r^i} (1 - \tau_{r, \tau}^i) - \frac{\tau_{d, \tau}}{\tau_d} + \frac{n_e}{n_n} \frac{1}{\tau_{a_2}} \right\} T_k ,$$

(19) 
$$\tau_{T} r_{k} T_{k} = - \varkappa k^{2} T_{k} - \left\{ F_{\text{VT}}(2 + \tau_{10,T}) + F_{\text{eTR}} + \frac{\tau_{\text{v}}^{1}(T)}{\tau_{10}} + \frac{\tau_{\text{v}}^{2}(T)}{\tau_{20}} \right\} T_{k} + \left\{ \frac{\tau_{\text{v}}^{1}(T_{1})}{\tau_{10}} + \frac{\tau_{\text{v}}^{2}(T_{1})}{\tau_{20}} \right\} T_{1k} + F_{\text{eTR}} n_{\text{e}k} + F_{\text{eTR}}(1 + r_{\text{TR},T_{\text{e}}}) T_{\text{e}k} ,$$

$$\begin{split} &(20) \qquad \tilde{\tau}_{\mathbf{v}}^{1} r_{k} \, T_{1k} = - \, z_{1} k^{2} \, T_{1k} \, + \\ &+ \left\{ F_{\mathbf{v}\mathbf{T}}(2 + \tau_{10,T}) - \tilde{F}_{\mathbf{e}\mathbf{v}}^{1} - F_{\mathbf{v}\mathbf{v}}(2 + \tau_{3,T}) - F_{\mathbf{p}\mathbf{h}\mathbf{v}}(2 - F_{\mathbf{p}\mathbf{h}\mathbf{v},T}) + \frac{\tau_{\mathbf{v}}^{1}(T)}{\tau_{10}} + \frac{\tau_{\mathbf{v}}^{2}(T)}{\tau_{20}} \right\} \, T_{+} - \\ &- \left\{ F_{\mathbf{v}\mathbf{v}} \, \tau_{3,T_{1}} - F_{\mathbf{p}\mathbf{h}\mathbf{v}} F_{\mathbf{p}\mathbf{h}\mathbf{v},T_{1}} + \frac{\tau_{\mathbf{v}}^{1}(T_{1})}{\tau_{10}} + \frac{\tau_{\mathbf{v}}^{2}(T_{1})}{\tau_{20}} + \frac{\tau_{\mathbf{v}}^{3}(T_{1})}{\tau_{3}} \right\} \, T_{1k} + \\ &+ \left\{ F_{\mathbf{p}\mathbf{h}\mathbf{v}} F_{\mathbf{p}\mathbf{h}\mathbf{v},T_{2}} + \frac{\tau_{\mathbf{v}}^{3}(T_{3})}{\tau_{2}} \right\} \, T_{0k} + \tilde{F}_{\mathbf{e}\mathbf{v}}^{1} n_{\mathbf{e}k} + \tilde{F}_{\mathbf{e}\mathbf{v}}^{1}(1 + r_{\mathbf{r}\mathbf{R},T_{\mathbf{e}}}) \, T_{\mathbf{e}k} + F_{\mathbf{p}\mathbf{h}\mathbf{v}} I_{k} \, , \end{split}$$

$$\begin{split} \tilde{\tau}_{\mathrm{v}}^{3} v_{k} T_{3k} &= \varkappa_{3} k^{2} T_{3k} + \left\{ -F_{\mathrm{ev}}^{3} + F_{\mathrm{vv}} (2 + \tau_{3,T}) \pm \frac{v_{3}}{v_{1}} F_{\mathrm{phv}} (2 - F_{\mathrm{phv},T}) \right\} T_{k} &\pm \\ &+ \left\{ F_{\mathrm{vv}} \tau_{3,T_{1}} \pm \frac{\tau_{\mathrm{v}}^{3} (T_{1})}{\tau_{3}} \pm \frac{v_{3}}{v_{1}} F_{\mathrm{phv}} F_{\mathrm{phv},T_{1}} \right\} T_{1k} - \left\{ F_{\mathrm{phv}} \frac{v_{3}}{v_{1}} F_{\mathrm{phv},T_{2}} \pm \frac{\tau_{\mathrm{v}}^{3} (T_{3})}{\tau_{3}} \right\} T_{3k} &\pm \\ &+ \tilde{F}_{\mathrm{ev}}^{3} n_{\mathrm{ek}} + \tilde{F}_{\mathrm{ev}}^{3} (1 + v_{\mathrm{TR},T_{\mathrm{e}}}) T_{\mathrm{ek}} - \frac{v_{3}}{v_{1}} F_{\mathrm{phv}} I_{k} \,, \end{split}$$

$$(22) T_{ek} = -\frac{2\cos^2 q}{\nu'_{u,T_e}} n_{ek} + \frac{2\sin^2 \varphi}{\nu'_{u,T_e}} T_k.$$

Here  $v_k = i\omega - i \boldsymbol{k} \cdot \boldsymbol{u}$  is the growth rate of the perturbation (propagating along the wave vector  $\boldsymbol{k}$  forming an angle  $\varphi$  with the static electric field  $\boldsymbol{E}$ ),  $\tau_{\rm d} = (n_{\rm n} k_{\rm d})^{-1}, \quad \tau_{\rm r}^{\rm e} = (n_{\rm p} k_{\rm r}^{\rm e})^{-1}, \quad \tau_{\rm a_1} = (n k_{\rm a_1})^{-1}, \quad \tau_{\rm a_2} = (n^2 k_{\rm a_2})^{-1}, \quad \tau_{\rm i} = (n k_{\rm i})^{-1} \quad {\rm and} \quad \tau_{\rm r}^{\rm i} = (n_{\rm n} k_{\rm r}^{\rm i})^{-1} \quad {\rm are} \quad {\rm the \ characteristic \ times \ of \ the \ processes \ leading \ to \ loss}$ 

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,

$$\left(\frac{1}{\tau_{a_1}}\right)$$
  $n_{a_k}$ 

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$$\left. \frac{1}{2} \right\} T_k +$$

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or generation of electrons,  $\tau_T = nC_pT/JE$  and  $\tau_{\rm v}^\alpha = n_\alpha C_{\rm v}^\alpha T_\alpha/JE$  are the characteristic times for heating the gas at the rate fixed by the electrical power JE deposited in the plasma,  $C_p$  is the specific heat at constant pressure and  $C_{\rm v}^{\rm v}$  the specific heat at constant volume of the  $\alpha$ -mode. Here and in the following, the logarythmic derivative of a quantity b with respect to a parameter c is indicated by  $b_{,c} \equiv \partial \ln b/\partial \ln c$ .  $F_{\rm err}$  is a measure of the fractional power transfer from electrons into transition-rotation excitation,  $F_{\rm vr} = \sum_{\alpha} F_{\rm rv}^\alpha$ ,  $F_{\rm vv} = \sum_{\alpha} F_{\rm vv}^\alpha$  and  $F_{\rm phv} = \sum_{\alpha} F_{\rm phv}^\alpha$ . I is the laser fieldi intensity. Finally,  $\varkappa_1$  and  $\varkappa_3$  are the vibrational thermal conductivities of the sets labelled 1 and 3, respectively.

The last equation of the system has been obtained by Haas and Nighan under the assumption of an instantaneous response of the electron energy to a local disturbance. In this equation  $v'_{u,\tau_e} = 1 + v_{u,\tau_e} - v_{m,\tau_e} \cos 2\varphi$ . It is worth noting that the density fluctuation  $n_k$  has been eliminated from the above system in virtue of the relation  $n_k = -T_k$  which holds in the initial phase of disturbance growth, when the pressure fluctuations are negligible.

For simplicity we have omitted the equation relating the laser intensity to the fluctuations of the plasma parameters and population inversion. This is justified by the circumstance that in the initial phase of the thermal instabilities, the gain of the medium can be considered to be constant. However, the coupling of intensity fluctuations with the medium density can lead to large-scale temporal oscillations (as predicted by Dreisen and Dykme (29) and observed by Yoder and Ahouse (30) in the output of high-power flowing laser devices) causing appreciable reduction in power output and optical-beam quality.

By simple algebra, the above system can be given the form

(23) 
$$\begin{cases} (a_{11} - \tau_r^c v_k) Y_{1k} + a_{12} Y_{2k} + A_{13} T_k = 0, \\ a_{21} Y_{1k} + (a_{22} - \tau_r^l v_k) Y_{2k} + A_{23} T_k = 0, \\ a_{31} Y_{1k} + (A_{33} - \tau_T v_k) T_k + a_{34} T_{1k} = 0, \\ a_{41} Y_{1k} + A_{43} T_k + (a_{44} - \tilde{\tau}_V^l v_k) T_{1k} + a_{45} = 0, \\ a_{51} Y_{1k} + A_{53} T_k + a_{54} T_{1k} + (a_{55} - \tilde{\tau}_V^3 v_k) T_{3k} = 0, \end{cases}$$

where  $Y_{1k} = n_{4k} + T_k$ ,  $Y_{2k} = n_{nk} + T_k$ ,  $A_{13} = a_{13} - a_{11} - a_{12}$ ,  $A_{23} = a_{23} - a_{21} - a_{22}$ ,  $A_{33} = a_{33} - a_{31}$ ,  $A_{43} = a_{43} - a_{41}$  and  $A_{53} = a_{53} - a_{51}$ . The coefficients are given in appendix A.

The growth rate  $r_k$  will be obtained by solving the characteristic equa-

<sup>(29)</sup> Y. Dreisen and A. M. Dykhne: JETP Lett., 19, 371 (1974).

<sup>(30)</sup> M. J. YODER and D. R. AHOUSE: Appl. Phys. Lett., 27, 673 (1979).

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tion  $D(r_k)=0$  of the above system. The search for a positive root has been carried out numerically. However, as a first approximation we can assume the instability to be either ionic or thermal. In the first case it amounts to putting  $T_k=T_{1k}=T_{3k}$  and replacing  $D(r_k)$  with a second-order polynomial  $D_{\text{ton}}(r_k)$ . The instability will be thermal when  $n_{ek}=n_{nk}=0$ . In such a case  $D(r_k)=0$  will reduce to a third-order polynomial. In virtue of this splitting, we can approximately write

(24) 
$$D(r_k) \simeq D_{ton}(r_k) D_{th}(r_k) = 0.$$

The resulting instability will be ionic or thermal according to whether the largest positive root of  $D_{\rm ion}=0$  is greater or smaller than the largest positive root of  $D_{\rm th}=0$ . The confidence in this approximation breaks down when the growth rates of thermal and ionic instabilities become comparable.

#### 6. - Results.

The system of equations (30) has been used to analyse the stability regions of a plasma produced in an e-beam preionized laser channel having a cross-section

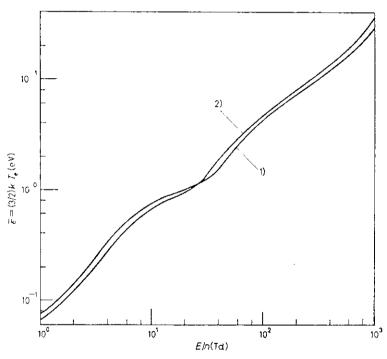


Fig. 1. – Calculated values of  $\bar{\epsilon} = \frac{3}{2}kT_e$  as a function of E/n. The curves 1) and 2) refer to the mixtures 1) and 2), respectively.

Fig. 2. =  $(n/W)^{(1)}$ 

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of  $10\cdot10$  cm. Two gas mixtures 1) 6:34:54:6 (CO<sub>2</sub>:N<sub>2</sub>:He:CO) and 2) 7.5:23: :62:7.5 entering the channel at a pressure of 66 Torr at 280 K have been examined. A power  $JE=8.7~\rm W/cm^3$  has been assumed as the factor producing the requested pumping of the preionized medium.

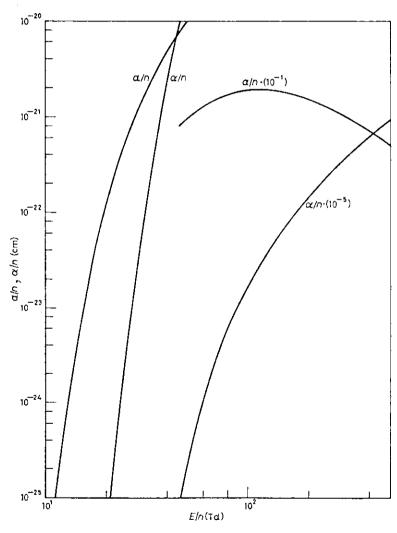


Fig. 2. – Calculated values of a/n and  $\alpha/n$  for mixture 1).  $k_a = (n/W)(\alpha/n)$  and  $k_1 = (n/W)(\alpha/n)$ .

The electron motion in the above mixtures, acted upon by the d.c. field of the sustainer, has been studied under conditions of special interest for the stability. Transport coefficients, *i.e.* electron drift velocity W, longitudinal and transverse diffusion coefficients  $D_{\rm L}$  and  $D_{\rm T}$  and ionization  $k_{\rm i}$  and attach-

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ment  $k_{\mathbf{a}_1}$  coefficients have been obtained as a function of E/n. The operating characteristics of the mixture have also been studied by calculating the fraction of electron energy which is lost by 1) elastic collisions, rotational excitation of  $CO_2$ ,  $N_2$ , CO and excitation  $(\widehat{F}_{\bullet v}^1)$  of the mixed (bending and symmetric stretching) modes of the  ${\rm CO_2};\ 2)$  excitation  $(\tilde{F}_{\rm ev}^3)$  of the asymmetric stretching mode of CO2 and the first eight vibrational levels of N2; 3) excitation of the first ten vibrational levels of CO  $(F_{ev}^{co})$ ; 4) electronic excitation of the gases of the mixtures and molecular dissociation of CO2, and 5) ionization of the gases of the mixture.

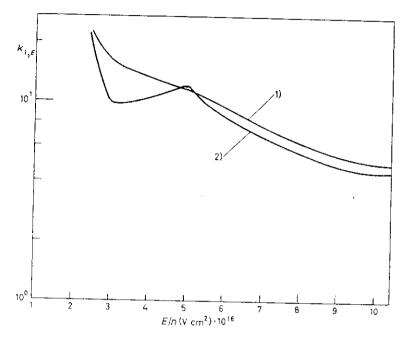


Fig. 3. - Logarithmic derivative  $k_{i,E}$  of the ionization coefficients for the two mixtures 1) and 2).

The calculations have been based on the cross-sections of Bulos and Phelps (81) for CO<sub>2</sub>, Engelhardt, Phelps and Risk (32) for N<sub>2</sub> and Land (33) for CO. For He, the cross-sections are the same used by Lowke, Phelps and IRWIN (34). This means that the electrons have been allowed to undergo 4 different kinds of elastic collisions and 48 different kinds of inelastic collisions (26 vibrational

Fig. 4. mixtures

(35) G. L. (1979).

<sup>(31)</sup> B. Bulos and A. V. Phelps: Phys. Rev. A, 14, 615 (1976).

<sup>(32)</sup> A. G. ENGHELHARDT, A. V. PHELPS and C. G. RISK: Phys. Rev. A, 135, 1566 (1964).

<sup>(33)</sup> J. E. LAND: J. Appl. Phys. (in print).

<sup>(34)</sup> J. J. LOWKE, A. V. PHELPS and B. W. IRWIN: J. Appl. Phys., 44, 4664 (1973).

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levels, 17 electronic levels, 4 ionizing processes and the dissociative process  $e+CO_2 \rightarrow CO+O^-$ ). The electrons have been also permitted to undergo rotational collisions, for which the continuous approximation has been used.

The details of the calculations have been discussed elsewhere (35). Here we will limit ourselves to presenting the calculated values of  $\bar{\varepsilon} = \frac{3}{2}kT_{\bullet}$  as a function of E/n (see fig. 1). The function  $T_{\bullet} = T_{\bullet}(E/n)$  is useful for expressing

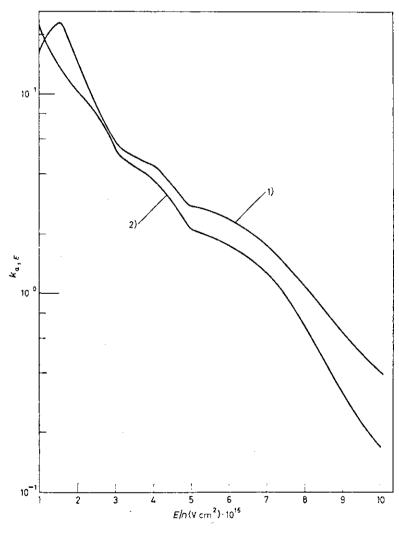


Fig. 4. – Logarithmic derivatives  $k_{a,E}$  of the attachment coefficients for the mixtures 1) and 2).

<sup>(35)</sup> G. L. Braglia, R. Bruzzese and G. L. Caraffini: Lett. Nuovo Cimento, 25, 139 (1979).

the logarythmic derivatives  $a_{,r_e}$  of quantities calculated as functions of E/n. In fig. 2 the ionization coefficient  $\alpha/n=(W/n)k_1$  and attachment coefficient  $a/n=(W/n)k_2$  are plotted for the mixture 1). In fig. 3 and 4 the derivatives of  $k_1$  and  $k_2$  are given for the two mixtures. The calculated functions  $F_{\rm ev}^{001}$ ,  $F_{\rm ev}^{010}$ ,  $F_{\rm ev}^{010}$ , are represented in fig. 5 for mixture 1).

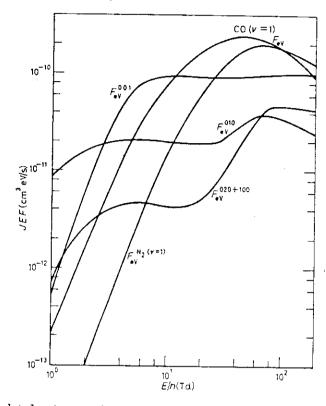


Fig. 5. – Calculated net power input to various vibrational levels of  ${\rm CO_2}$ ,  ${\rm N_2}$  and  ${\rm CO}$  as a function of E/n for mixture 1).

The translational and vibrational temperatures along the channel have been calculated by using a numerical code LEBINT which has been developed for a four-component e-beam preionized laser mixture. The transport coefficients and the excitation rates of the vibrational levels used in the LEBINT code are a best fitting of those obtained with our Boltzmann code and those reported by Kurzius and Thoenes (36). The plasma recombination rate has been calculated by using some experimental data concerning the transconduc-

tance of a found in relaser mixto

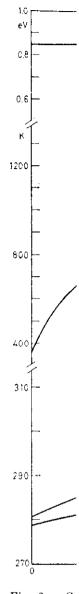


Fig. 6. - Ca tures along

<sup>(36)</sup> J. Thoenes and S. C. Kurzius: EDL Performance Model, Part III, Report No. RG-CR-75-2, Lockheed Missiles and Space Company (1976).

<sup>(37)</sup> S. MAI Cimento B,

ons of E/n. t coefficient derivatives actions  $F_{ev}^{001}$ ,

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, N<sub>2</sub> and CO

annel have developed port coeffie LEBINT and those n rate has

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tance of a typical EDCL device. A detailed account of this program can be found in ref. (57). The results of the simulation are plotted in fig. 6 for the laser mixture 1).

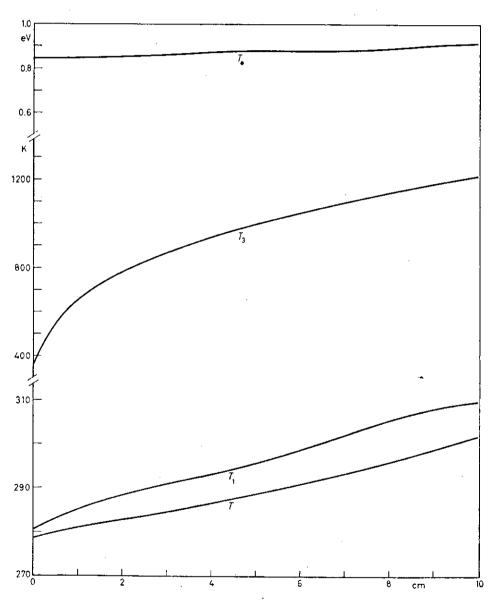


Fig. 6. – Calculated electronic  $(T_e)$ , vibrational  $(T_3, T_1)$  and translational (T) temperatures along the laser channel for mixture 1).

<sup>(37)</sup> S. MARTELLUCCI, J. QUARTIERI, G. MASTROCINQUE and S. SOLIMENO: Nuovo Cimento B, 54, 99 (1979).

The time constants  $\tau_{\mathbf{r}}$ ,  $\tilde{\tau}_{\mathbf{v}}^{1}$  and  $\tilde{\tau}_{\mathbf{v}}^{4}$  have been calculated (see fig. 7) by using these temperature profiles along the laser channel.

The roots of the instability dispersion equation  $D(r_k) = 0$  have been calculated for different values of k. In so doing, it has also been verified that for  $k^{-1} = l > 0.3$  cm the roots are practically independent of k, as pointed out by Nighan.

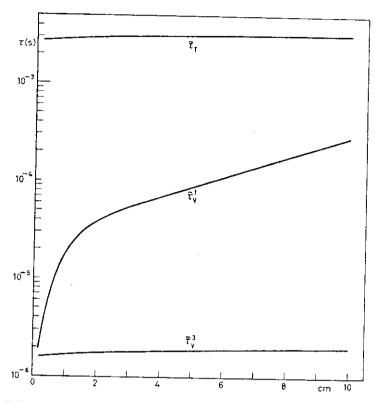


Fig. 7. – Calculated time constants  $\tau_T$ ,  $\bar{\tau}_V^1$  and  $\bar{\tau}_V^3$  along the laser channel for mixture 1).

In general, only one positive root has been found. The unstable mode has resulted to be ionic for the two mixtures 1) and 2) under the referred conditions. The relative growth rate has been shown to be strongly dependent on the concentration of negative ions. This is clearly displayed in fig. 8, where calculated values of  $\nu_k$  for different ratios  $n_n/n_e$  have been plotted.

The fluctuations  $T_{ik}$  of the vibrational temperatures and of the electronic density  $n_{ek}$  have been calculated for  $n_{ek}/n_{e}=10^{-1}$  by putting  $T_{k}=1$ . The plots of fig. 9 confirm the relative high value of the electronic fluctuations with respect to the translational temperature, as requested by the ionic character

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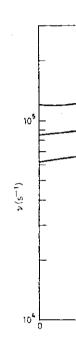


Fig. 8. - ( ferent conc

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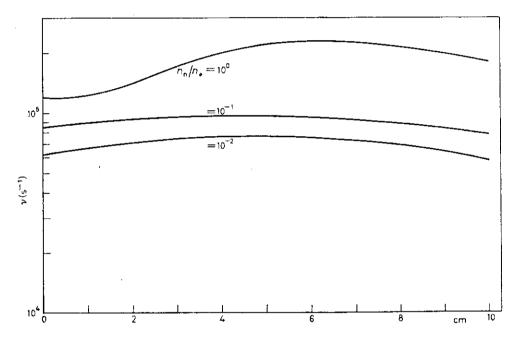


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. 7) by using

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proves the need for decoupling the fluctuations of the lower laser level temperature from the translational one. This provides clear evidence to the opportunity of improving the vibrational kinetic model of the Haas-Nighan theory. pointed out Due to the rapid increase of  $T_1$  with respect to T during the instability onset, the fluctuation of the laser upper-level temperature can grow so strong as to overcome that of the electronic density. This, however, does not destroy the ionic character of the instability which has been confirmed by solving for the approximate dispersion equation  $D_{ion}(\nu_k) = 0$ . In fact, the growth rates plotted in fig. 10 agree with those of fig. 8.



of the instability mode. On the other hand, the relatively high value of  $T_{1k}$ 

Fig. 8. - Calculated growth rate of ionic instability along the laser channel for different concentration of negative ions in mixture 1).

For mixture 1) the approximate dispersion equation  $D_{tb}(r_k) = 0$  has also been tested. The relative values of  $r_k$  are presented in fig. 11 for different laser intensities. The observed drastic reduction of  $r_k$  for  $F_{\rm ph}=0.1$  confirms the conjecture that lasing of the active plasma produces a stabilizing effect on the thermal modes. As a consequence, allowing the laser intensity to grow with the electric power deposited in the plasma, the thermal collapse of the discharge can be efficiently contrasted.

To complete the analysis of the ionic instabilities, the derivatives of  $k_*$  vs.  $k_*$ have been calculated and represented in fig. 12 for the two laser mixtures.

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le mode has t conditions. lent on the where cal-

e electronic  $_{k}=1$ . The fluctuations ic character

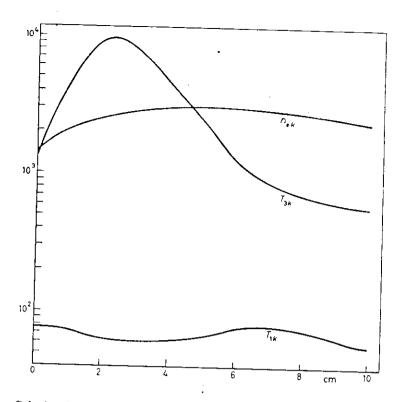


Fig. 9. – Calculated fluctuations of the vibrational temperatures  $(T_{1k}$  and  $T_{3k})$  and of the electronic density  $n_{nk}$  for  $n_n/n_e=10^{-1}$  for mixture 1).

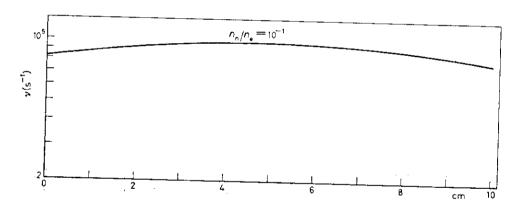


Fig. 10. – Growth rate of ionic instability following from the reduced dispersion equation  $D_{\text{lon}}(\nu_k)=0$ . Note the good agreement with the corresponding curve of fig. 8.

Fig. 11. - different in  $F_{\rm ph}$  rises to

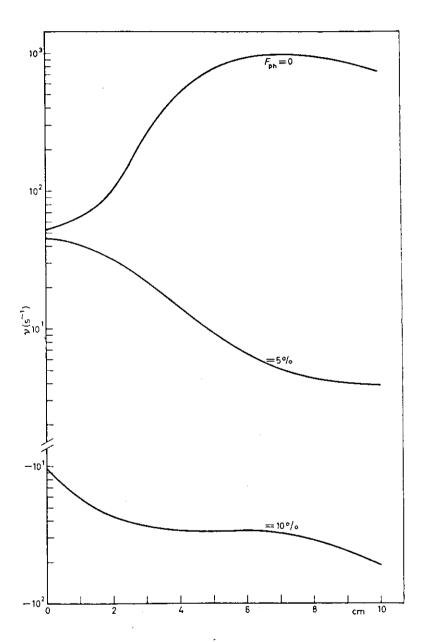


Fig. 11. – Calculated growth rates of thermal instability along the laser channel for different intensities  $(F_{\rm ph})$  of the laser beam. Note the dramatic decrease of  $\nu$  when  $F_{\rm ph}$  rises to 10% of JE.

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 $\operatorname{ad} T_{3k}$ ) and

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ersion equave of fig. 8.

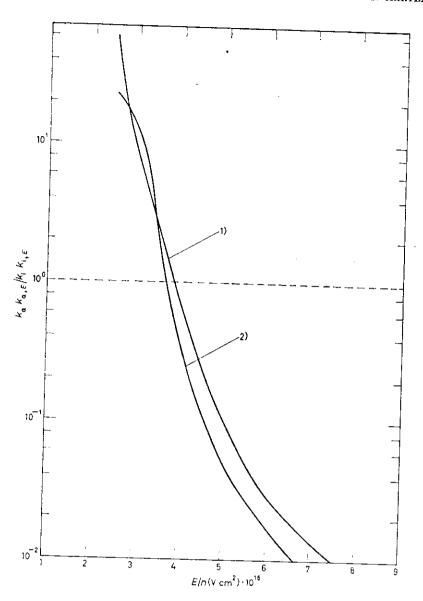


Fig. 12. – Instability region obtained by using the Nighan simplified criterion  $k_{\mathbf{a}}k_{\mathbf{a},\mathbf{z}}^*/k_{\mathbf{i}}k_{\mathbf{i},\mathbf{z}} = \partial k_{\mathbf{a}}/\partial k_{\mathbf{i}} > 1$ , for the two mixtures examined.

As shown by Nighan, Wiegand and Haas, the attachment must dominate over the ionization during an unstable fluctuation. This leads to the criterion  $\partial k_*/\partial k_i > 1$  for formation of striations. By using the curves of fig. 12, the occurrence of striations can be excluded for E/n exceeding 40 townsend.

#### 7. - Concl

In the e-beam-su discussion Nighan as considered

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APPEND

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$$a_{11} = - \left\{1\right\}$$

$$a_{12} = \frac{n_n}{n_\bullet} \frac{\tau_\tau^e}{\tau_d}$$

$$a_{13} = -$$

$$a_{14}=0,$$

$$a_{15}=0,$$

#### 7. - Conclusions.

In the present paper a general model of plasma instability in high-power e-beam-sustained CO<sub>2</sub> laser amplifiers is presented together with a detailed discussion of some experimental cases. Our model improves that of Haas-Nighan as the various degrees of vibrational freedom and the laser field are considered.

The model has been used to test the degrees of thermal and ionic instabilities of an EDCL device using 4-component gas mixtures (CO,-N,-He-CO). The mixtures 1) 6:34:54:6 and 2) 7.5:23:62:7.5 have been examined.

The fifth-order dispersion equation resulting from the linearization of the fluid dynamic and kinetic equations is found to have only one positive (i.e. unstable) root. For the above laser parameters, this unstable mode has resulted to be ionic for both mixtures.

The stabilizing effect of the laser beam intensity has also been assessed, thus showing a dramatic reduction of the growth rate of thermal instabilities. Consequently, there is enough evidence for considering thermal instabilities of secondary relevance with respect to the formation of moving strictions in EDCL lasers.

It is a pleasure to thank W. NIGHAN for calling the attention on some of the quoted literature and for helpful discussions with one of us (SS). Also numerous discussions and comments by Profs. F. PANDARESE, O. SVELTO and A. Sona are much appreciated. The temperature profiles were provided by Dr. J. QUARTIERI whose co-operation was of much help.

#### APPENDIX A

The coefficients of the stability system (30) are

$$\begin{split} a_{11} &= -\left\{1 + \frac{n_e}{n_p} + \tau_r^e \left(\frac{1}{\tau_{a_1}} + \frac{1}{\tau_{a_2}} - \frac{1}{\tau_i}\right) - \frac{2\cos^2\varphi}{\nu'_{u,\tau_e}} \tau_r^e \left(\frac{\tau_{r,\tau_e}^e}{\tau_r^e} + \frac{\tau_{a_1,\tau_e}}{\tau_{a_1}} + \frac{\tau_{a_1,\tau_e}}{\tau_{a_2}} - \frac{\tau_{i,\tau_e}}{\tau_i}\right)\right\},\\ a_{12} &= \frac{n_n}{n_e} \frac{\tau_r^e}{\tau_d} - \frac{n_n}{n_p},\\ a_{13} &= -\left\{1 + \frac{n_n}{n_e} \frac{\tau_r^e}{\tau_d} \tau_{d,r} - \frac{\tau_r^e}{\tau_{a_2}} + \frac{2\sin^2\varphi}{\nu'_{u,\tau_e}} \tau_r^e \left(\frac{\tau_{r,\tau_e}^e}{\tau_r^e} + \frac{\tau_{a_1,\tau_e}}{\tau_{a_1}} + \frac{\tau_{a_1,\tau_e}}{\tau_{a_2}} - \frac{\tau_{i,\tau_e}}{\tau_i}\right)\right\},\\ a_{14} &= 0,\\ a_{15} &= 0, \end{split}$$

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$$a_{21} = -\frac{n_e}{n_p} + \frac{n_e}{n_n} \tau_r^1 \left\{ \frac{1}{\tau_{a_1}} + \frac{1}{\tau_{a_1}} - \frac{2\cos^2 \varphi}{\nu'_{u,\tau_e}} \left( \frac{\tau_{a_1,\tau_e}}{\tau_{a_1}} + \frac{\tau_{a_2,\tau_e}}{\tau_{a_2}} \right) \right\},$$

$$a_{22} = -\frac{n_n}{n_p} + \frac{n_e}{n_n} \tau_r^1 \left( \frac{1}{\tau_{a_1}} + \frac{1}{\tau_{a_2}} \right),$$

$$a_{22} = -1 - \tau_{a_2}^1 - \frac{\tau_r^1}{\tau_{a_2}} - \frac{n_e}{\tau_{a_2}} + \frac{1}{\tau_{a_2}} \left( \frac{1}{\tau_{a_1,\tau_e}} + \frac{2\sin^2 \varphi}{\tau_{a_2,\tau_e}} + \frac{\tau_{a_2,\tau_e}}{\tau_{a_2,\tau_e}} \right)$$

$$\begin{aligned} a_{23} &= -1 - \tau_{r,T}^{1} - \frac{\tau_{r}^{1}}{\tau_{d}} \tau_{d,T} - \frac{n_{o}}{n_{n}} \tau_{r}^{1} \left\{ \frac{1}{\tau_{a_{1}}} + \frac{2 \sin^{2} \varphi}{\nu_{u,T_{o}}'} \left( \frac{\tau_{a_{1},T_{o}}}{\tau_{a_{1}}} + \frac{\tau_{a_{2},T_{o}}}{\tau_{a_{1}}} \right) \right\}, \\ a_{24} &= 0. \end{aligned}$$

$$a_{24}=0,$$

$$a_{25}=0$$
,

$$a_{\rm al} = F_{\rm eTR} \left\{ 1 - \frac{2\cos^2 \varphi}{v'_{u,T_{\rm e}}} (1 + v_{\rm TR}, \tau_{\rm e}) \right\},$$

$$a_{32}=0,$$

$$a_{33} = -\left\{\frac{k}{l^2} + F_{\text{VT}}(2 + \tau_{10,T}) + F_{\text{eTR}} + \frac{\tau_{\text{v}}^1(T)}{\tau_{10}} + \frac{\tau_{\text{v}}^2(T)}{\tau_{20}} - F_{\text{eTR}} \frac{2\sin^2\varphi}{\nu_{\text{u},\tau_e}'} (1 + \nu_{\text{TR},\tau_e})\right\},\,$$

$$a_{34} = \frac{\tau_{\rm v}^1(T_1)}{\tau_{10}} + \frac{\tau_{\rm v}^2(T_1)}{\tau_{20}} \,,$$

$$a_{35}=0,$$

$$a_{\rm si} = \tilde{F}_{\rm ev}^1 \left\{ 1 - \frac{2 \, \cos^2 \varphi}{v_{\rm u,r_e}'} (1 \, + \, v_{\rm TR,r_e}) \right\}, \label{eq:asimple}$$

$$a_{42} = 0$$

$$\begin{split} a_{43} &= F_{\rm VT}(2\,+\,\tau_{10,\rm f}) - \tilde{F}_{\rm ev}^1 - F_{\rm vv}(2\,+\,\tau_{\rm 3,\rm f}) - F_{\rm phv}(2\,-\,F_{\rm phv},\rm f}) \,+ \\ &\quad + \frac{\tau_{\rm v}^1(T)}{\tau_{10}} + \frac{\tau_{\rm v}^2(T)}{\tau_{20}} + \tilde{F}_{\rm ev}^1 \,\frac{2\,\sin^2\varphi}{\nu_{\rm u,f,e}'} \,(1\,+\,\nu_{\rm TR,f_e}) \,, \end{split}$$

$$a_{44} = -\left\{ \frac{k_1}{l^2} + F_{vv}\tau_{3,\tau_1} - F_{phv}F_{phv,\tau_1} + \frac{\tau_v^1(T_1)}{\tau_{10}} + \frac{\tau_v^2(T_1)}{\tau_{20}} + \frac{\tau_v^3(T_1)}{\tau_3} \right\},\,$$

$$a_{45} = F_{
m phv} F_{
m phv, au_3} + rac{ au_{
m v}^3(T_3)}{ au_3} \, ,$$

$$a_{\rm sI} = \tilde{F}_{\rm ev}^{\rm a} \left\{ 1 - \frac{2 \cos^2 \varphi}{v_{\rm u, \tau_e}'} (1 + v_{\rm TR, \tau_e}) \right\},$$

$$a_{52} = 0$$

$$a_{\rm 53} = -\,\tilde{F}_{\rm ev}^3 + F_{\rm vv}(2\,+\,\tau_{\rm 3,\it t}) \,+\, \frac{v_{\rm 3}}{v_{\rm 1}}\,F_{\rm phv}(2\,-\,F_{\rm phv,\it t}) \,+\,\tilde{F}_{\rm ev}^3\,\frac{2\,\sin^2\varphi}{v_{\rm u,\it t_e}'}(1\,+\,v_{\rm tr,\it t_e})\;, \label{eq:a53}$$

$$a_{\rm 54} = F_{\rm vv} \, \tau_{\rm 3,r_1} + \frac{\tau_{\rm v}^{\rm I}(T_{\rm 1})}{\tau_{\rm 3}} + \frac{\nu_{\rm 3}}{\nu_{\rm 1}} F_{\rm phv} F_{\rm phv,r_1} \,,$$

$$a_{55} = -\left\{ \frac{k_3}{l^2} + F_{
m phv} \frac{v_3}{v_1} F_{
m phv, r_4} + \frac{\tau_{
m v}^3(T_3)}{\tau_3} \right\}.$$

APPENDI

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where (3.37)

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(B.9)

APPENDIX B

#### Logarithmic derivatives of some factors appearing in the stability system (23).

Assuming for  $k_i^i$  a dependence on T of the form suggested by the SSH theory

(B.1) 
$$k_{j}^{i}(T) = A_{j}^{i} \exp\left[-B_{j}^{i} T^{-1}\right],$$

we have

(B.2) 
$$\tau_{10,r} = \tau_{20,r} = -n_{,r} + \frac{\sum_{j} X_{j} k_{j,r}^{j}}{\sum_{j} X_{j} k_{j}^{j}},$$

where (3.37)

(B.3) 
$$\begin{cases} k_{20,T}^{\text{co}_{2}} \simeq 1.270 \cdot 10^{-2} (\text{KT}) , & 1.067 \cdot 10^{-2} (\text{SSH}) , \\ k_{20,T}^{\text{Nc}} \simeq 1.270 \cdot 10^{-2} (\text{KT}) , & 1.067 \cdot 10^{-2} (\text{SSH}) , \\ k_{20,T}^{\text{co}} \simeq 0.330 \cdot 10^{-2} (\text{KT}) , & 1.067 \cdot 10^{-2} (\text{SSH}) , \\ k_{20,T}^{\text{He}} \simeq 0.666 \cdot 10^{-2} (\text{KT}) , & 0.747 \cdot 10^{-2} (\text{SSH}) . \end{cases}$$

KT stands for the initials of the authors of ref. (37). Due to the smallness of these factors we can put

(B.4) 
$$\tau_{10,T} = \tau_{20,T} \simeq -n_{,T} = -1.$$

Analogously, for  $\tau_{3,\tau_1}$  and  $\tau_{3,\tau}$ , we have

(B.5) 
$$\tau_{s,r} \simeq -\frac{3}{2} + \frac{\Delta E}{kT},$$

(B.6) 
$$\tau_{3,T_1} \simeq \frac{3000}{T_1} \exp\left[\frac{423}{T} - \frac{3000}{T_1}\right].$$

Differentiating eq. (22) yields

(B.7) 
$$F_{\rm phv,T} \simeq \frac{hcBJ(J+1)}{kT} \simeq 1.88 \cdot 10^{-16}$$
,

$$(\mathrm{B.8}) \qquad F_{\mathrm{phv},r_1} = -\tilde{Q}_{\mathrm{I},r_1} - \left\{ \exp\left[\theta_1 - \theta_3\right] \frac{P(J)\,\Theta_J}{P(J+1)\,\Theta_{J+1}} - 1 \right\}^{-1} \simeq -4.3\cdot 10^{-2} \; ,$$

(B.9) 
$$F_{\text{phv},\tau_2} = \tilde{Q}_{3,\tau_2} - \left\{ \exp\left[-\theta_1 + \theta_3\right] \frac{P(J+1)\theta_{J+1}}{P(J)\theta_J} - 1 \right\}^{-1} \simeq 1.17$$
.

 $(1+\nu_{\text{TR},\tau_{\bullet}})$ ,

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 $(1 + \nu_{\text{TR},r_e})$ ,

 $-\nu_{\mathrm{TR},T_{\bullet}})$ ,

#### • RIASSUNTO

Nel presente lavoro si presenta un modello generale delle instabilità di plasma in un laser a  $\mathrm{CO}_2$  di alta potenza, preionizzato a fascio elettronico, insieme con una dettagliata discussione di alcuni casi sperimentali. Il presente modello è un'estensione di quello aviluppato da Haas e poi da Nighan, dal momento che si tiene esplicitamente conto dei vari gradi di libertà vibrazionali e dell'effetto del campo laser. Il suddetto modello è stato usato per analizzare il grado d'instabilità ionica e termica in un laser del tipo EDCL utilizzante miscele gassose quaternarie ( $\mathrm{CO}_2\text{-N}_2\text{-He-CO}$ ). Si è messo chiaramente in luce l'effetto stabilizzante del fascio laser, mostrando la netta riduzione del ritmo di crescita delle instabilità di tipo termico all'aumentare dell'intensità del fascio.

Ионные и тепловые неустойчивости в EDCL устройствах, с преднонизацией  ${\rm CO_2}$  электронным пучком, в присутствии лазерного луча.

Резюме (\*). — В этой работе предлагается общая модель плазменных неустойчивостей в  $CO_2$  лазерных усилителях и подробно обсуждаются некоторые экспериментальные случаи. Эта модель улучшает модель Хааса-Нигана, допуская в явном виде различные степени вибрационной свободы и учитывая лазерное поле. Предложенная модель используется для проверки степени тепловой и ионной неустойчивости EDCL устройства, использующего четырех-компонентные газовые смеси  $(CO_2-N_2-He-CO)$ . Стабилизирующий эффект интенсивности лазерного пучка приводит к существенному уменьшению скорости роста тепловых неустойчивостей.

(\*) Переведено редакцией.

