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Kinetics of a CO, Nuclear Pumped Laser

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H. A. Hassan*
North Carolina State University, Raleigh, N.C.

Abstract

IRECT nuclear pumping was demonstrated at a number of laboratories using molecular and atomic systems with one notable exception, i.e., CO₂. This work is being undertaken with the objective of explaining the mystery surrounding the direct nuclear pumping of CO₂. To achieve this objective, a detailed kinetic model which incorporates all important reactions in a ³He-N₂-CO₂ mixture is formulated. The results show that for mixtures typical of those employed in electric discharge systems, the gain coefficients are such that lasing is not expected to take place. Moreover, for conditions representative of current nuclear-pumped laser experiments, concentrations of CO₂ in the 1-3% range are optimum.

Contents

The experimental setup representative of a nuclear-pumped laser by a volume source of fission fragments consists of a tube filled with fissionable material and a lasing medium surrounded by a moderator and placed in a fast burst reactor. When the thermalized neutrons interact with ³He, high-energy protons and tritons are formed and these, together with the primary and secondary electrons, ionize and excite the gas. The electrons are essentially thermal but non-Maxwellian¹; thus, the resulting plasma is recombination dominated. This implies that, in general, the dominant pumping mechanism is collisional recombination. However, when one deals with molecular lasers such as CO₂, electron excitation is also important. Thus, both mechanisms must be considered simultaneously.

The dominant substance in the system is assumed to be He and, as such, the energy of the fission fragments is deposited first into He in the form of ionization and excitation. The production rates of the ionized state He + and the excited state He* are derived from the power density released by the ³He (n,p) ³H reaction²

$$P_d = 9.3 \times 10^{-18} p\phi_0 X_{\text{He}} \text{ (kW/cm}^3)$$
 (1)

and the energy required to create an excited or an ionized state, i.e., the w values. In Eq. (1), p is the pressure in atmospheres, ϕ_0 is the flux (N/cm² s) of thermal neutrons, and $X_{\rm He}$ is the He fraction. Equation (1) is valid for tubes of diameters of the order of 2-3 cm and pressures of the order of 1 atm.

The energy deposited in He is transferred to CO_2 and N_2 by charge transfer, Penning ionization, and recombination. The kinetic model employed incorporates all of the above processes together with direct electron excitation, V-V and V-T energy transfer. Charge transfer reactions involving ions with large recombination energies, such as He^+ , and

polyatomic molecules tend to produce mainly dissociative ion products. Dimer ions, such as He₂+which is formed according to the reaction

$$He^{+} + 2He \rightarrow He^{+}_{2} + He$$
 (2)

have somewhat lower recombination energies and therefore do not cause as much dissociative ionization. On the other hand, Penning ionization tends to populate all the energetically accessible electronic molecular energy levels and, consequently, generates emission from a variety of electronic states and vibrational rotational sublevels within a given state.³

Although rates involving charge transfer reactions are known, in most cases the reaction products are not. Parker et al. 4 indicate that the fragment ion produced in highest abundance is expected to be the one whose appearance potential (AP) is nearest to the recombination energy (RE) of the bombarding ion. In other words, a process for which the energy defect $\Delta E = RE - AP$ is nearest to zero should predominate. This suggestion was employed here for those reactions where measurements of reaction products are not available. Similar uncertainties exist when one considers recombination products. Because one of the objectives of this work is to determine why nuclear pumping of CO_2 has not been possible, conservative assumptions regarding the neutral stabilized recombination of N_2^+ and CO_2^+ are employed. Thus, it is assumed that

$$N_2^+ + e + M - N_2 + M$$
 (3)

$$CO_2^+ + e + M \rightarrow CO_2^* + M$$
 (4)

In Eq. (3), N_2 is assumed to be the ground state, while in Eq. (4), CO_2 is assumed to be the lower laser level. The latter assumption is prompted by the observation that transitions in CO_2^+ involved mostly symmetric and bending modes.

Calculations indicated that the rates for direct electron excitation of the lower laser level were higher than those for the upper level. When this is combined with Eq. (4) and following assumption together with the observation that CO_2^+ happened to be the dominant ion in the system, one concludes that direct nuclear pumping of CO_2 is not possible. However, if N_2 is present, then the direct electron excitation of N_2 exceeds that for the lower laser level. The V-V energy transfer

$$CO_2 + N_2(V=1) \neq CO_2(001) + N_2$$
 (5)

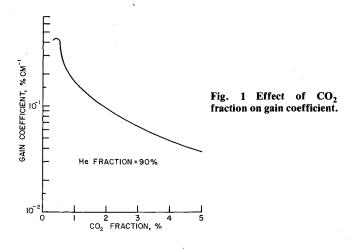
is responsible for pumping the upper laser level. Even when one ignores the role of direct electron excitation, one finds that the concentration of $N_2(V=1)$, resulting from the recombination of the excited nitrogen atoms, is such that lasing is possible under certain conditions.

Calculations were carried out for various mixtures, pressures, and fluxes for a single cavity 60 cm long, mirror reflectivities of 1.0 and 0.99, and radii of curvature of 2 m. Figures 1 and 2 show the effects of CO_2 on gain and power output for a total pressure of 1 atm, a neutron flux of 3×10^{16} N/cm² s, and a temperature of 300 K. It is seen that there is an optimum concentration where the gain or power output is

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^{*}Professor. Associate Fellow AIAA.



optimum. This can be traced to the fact that the upper laser level increases with CO_2 concentration and with N_2 (V=1). Because the total fraction of CO_2 and N_2 is fixed for this calculation, an optimum must exist. Calculations indicate also that for a given mixture there is an optimum pressure for which power output is maximum; in addition, for a given cavity there is a pressure above which the power output is zero.

In conclusion, the analysis indicates that a nuclear-pumped laser may operate under conditions of low total pressure and low percentages of CO_2 and N_2 . Under these conditions, the CO_2 laser is inefficient and is not scaleable. This behavior, which is in contrast to electric discharge CO_2 lasers, stems from the fact that the excitation mechanisms in both cases are quite different.

Acknowledgment

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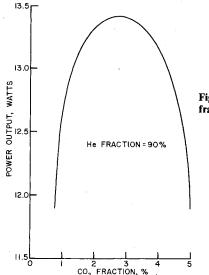


Fig. 2 Effect of CO₂ fraction on power output.

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