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# LASER EMISSIONS FROM CO<sub>2</sub> VIBRATIONAL TRANSITIONS IN A LOW TEMPERATURE SUPERSONIC FLOW EXCITED BY A PULSED ELECTRON BEAM STABILIZED DISCHARGE

B. Fontaine, B. Forestier, P. Gross and E.M. Koudriavtsev\*.

\**Institute of Fluid Mechanics, Aix-Marseille University, 1, rue Honnorat, 13003 Marseille, France.*  
 \**P.N. Lebedev Institute of Physics, Academy of Sciences of U.S.S.R., Moscow, U.S.S.R..*

**Résumé.**— Des émissions laser infrarouge de grande puissance en régime d'impulsions longues ont été obtenues au moyen d'un dispositif permettant l'excitation par une décharge stabilisée par faisceau d'électrons d'un milieu gazeux à basse température et densité élevée en écoulement supersonique ( $\rho < 2$  amagats,  $T \approx 60 - 150$  K). Une émission laser à  $\lambda = 10,6 \mu$  a été obtenue lorsque une cavité résonnante était placée soit dans la zone d'excitation, soit à 3 cm en aval de celle-ci. Dans le cas de mélanges Ar/CO<sub>2</sub>, Ar/CO<sub>2</sub>/H<sub>2</sub>, He/CO<sub>2</sub> et He/CO<sub>2</sub>/N<sub>2</sub>, la puissance et l'énergie de l'émission laser étaient très notablement plus élevées à basse température. L'évolution dans le temps de l'émission laser était très différente à basse température et à température ambiante. Lorsque la cavité résonnante était placée en aval, en dehors de la décharge, une émission laser infrarouge était observée pendant plusieurs dizaines de microsecondes pour des mélanges He/CO<sub>2</sub>/N<sub>2</sub> en écoulement supersonique. Des résultats expérimentaux sont présentés en ce qui concerne les caractéristiques de l'émission laser à  $10,6 \mu$  et la recherche d'émissions laser à partir de la molécule CO<sub>2</sub>, à de plus grandes longueurs d'onde généralement favorisées par de très basses températures de translation ( $14 \mu$  (100-010),  $16 \mu$  (020-010);  $18 \mu$  (03 0-10 0)). Les implications des résultats de l'I.M.F.M. pour le développement de sources laser infrarouges de puissance moyenne et de taux de répétition élevés, particulièrement utiles en photochimie, sont discutées.

**Abstract.**— High power long pulse infrared laser emission has been achieved on CO<sub>2</sub> molecule with the high density and very low temperature supersonic flow-electron beam-stabilized discharge excitation device developed at I.M.F.M. ( $\rho < 2$  amagats,  $T \approx 70 - 150$  K). Laser emission at  $\lambda = 10.6 \mu$  has been achieved for a resonant cavity set at the discharge location and also 3 cm downstream of the discharge location. With Ar/CO<sub>2</sub>, Ar/CO<sub>2</sub>/H<sub>2</sub>, He/CO<sub>2</sub>, and He/CO<sub>2</sub>/N<sub>2</sub> mixtures, lasing energy and power were several times higher at low temperature when experiments were performed at the discharge location. Time variation of laser power was very different compared to the room temperature, no flow, case for the same density and excitation energy. When the resonant cavity was set 3 cm downstream, outside of the discharge, lasing in the infrared was observed during several tens of microseconds for He/CO<sub>2</sub>/N<sub>2</sub> mixtures in supersonic flow. Experimental results are presented for both characterisation of  $10.6 \mu$  laser emissions and research of lasing at longer wavelength on CO<sub>2</sub> molecule, on the  $14 \mu$  (100-010) and  $16 \mu$  (020-010) cascade transitions and  $18.4 \mu$  (03 0-10 0) transition which are generally favored by very low translational temperature. Implications of I.M.F.M. results for development of high average power high repetition rate infrared laser sources useful for photochemistry are discussed.

## INTRODUCTION

The CO<sub>2</sub> vibrational flow lasers have shown their ability to deliver high average power at  $10.6 \mu$  and applications of these lasers are already numerous in particular in the Energetics field. Fluid Dynamics Techniques associated with electrical excitation are of high interest to achieve such high average power infrared laser systems.

Recently in several laboratories lasing has been achieved on different other vibrational transitions of CO<sub>2</sub> molecule, in particular on the (100-010) and (020-010) cascade transitions by

saturation of the 100 and 020 vibrational levels following lasing at  $10.6 \mu$  and  $9.4 \mu$  <sup>(1),(2)</sup> and on the (030-100) transition <sup>(3)</sup> respectively at  $\lambda = 14 \mu$ ,  $16 \mu$  and  $18.4 \mu$ . These wavelengths are interesting for laser photochemistry applications and the  $16 \mu$  wavelength has been recently proposed as a coherent source for isotopic separation. Different methods have been used to obtain population inversion including purely gas dynamics means (heating at thermodynamic equilibrium followed by very fast adiabatic expansion) <sup>(1)</sup> or direct electrical

excitation of a low pressure, low temperature, gas mixture without flow<sup>(2)</sup>. In all these experiments, active medium was at very low temperature to maintain a low population of the laser transition lower level, and at low pressure, to avoid vibrational deactivation. This last characteristic limits strongly the specific laser power which may be extracted from the active medium.

Recent results on CO<sub>2</sub> molecule kinetics<sup>(4)</sup> shows that conditions may exist where lasing could be achieved on various CO<sub>2</sub> vibrational transitions at higher pressure if the active medium is maintained at very low temperature and efficiently and rapidly excited. A pulsed U.V. preionized or e-beam stabilized discharge would be a possible method of excitation. Lasing at higher pressure would permit : a) higher laser average power on the pulsed high repetition rate mode and b) higher tunability of the laser wavelength following pressure broadening of the gain bandwidth. Moreover conditions of very low temperature and quite high density may also favor, as predicted by rough calculations, the already very efficient CO<sub>2</sub> laser at  $\lambda = 10.6\mu$  compared to room temperature by an increase of gain and a decrease of desactivation losses when a high density medium is excited by an electric discharge. On Fig. 1 is shown a vibrational energy level diagram of the CO<sub>2</sub>/N<sub>2</sub> vibrational system. Various possible CO<sub>2</sub> vibrational laser transitions are indicated on this figure.

Cooling of the gas by means of a supersonic expansion could lead to high repetition rate laser systems as the heat and waste products are very rapidly carried away by the supersonic flow together with the aerodynamic disturbances induced by energy deposition. Supersonic flow may represent also a way to control the relaxation of the lower laser level while maintaining nearly frozen the

population of the upper level by simply shifting the cavity location a few centimeters downstream the discharge position owing to the various relaxation rate constants.

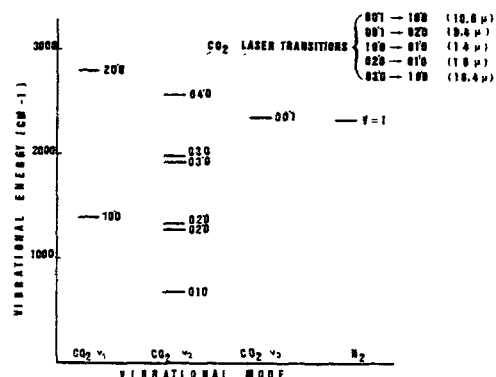


Fig.1 Vibrational energy level diagram for the CO<sub>2</sub>/N<sub>2</sub> molecular system (partial)

Associated with supersonic flow and adiabatic cooling which permits moreover to reach temperatures not easily achievable with thermal exchangers, the use of an electron beam stabilized discharge is a very efficient method for vibrational level excitation and could allow development of high average power high efficiency laser systems at various infrared wavelengths.

The aim of this paper is to present results of preliminary experiments on CO<sub>2</sub> vibrational laser emissions performed with the supersonic flow - electron beam stabilized discharge excitation device developed at I.M.F.M. for excimer laser studies. This device which has permitted to realize the first supersonic flow excimer laser<sup>(5)</sup> and to deliver laser specific energy and power of 1 J/ℓ and 2 MW/ℓ respectively with over 2.5 % intrinsic efficiency from XeCl laser system at very low temperature in supersonic flow<sup>(6)</sup> has been slightly modified for the infrared laser experiments.

#### EXPERIMENTAL SET-UP

The basic experimental apparatus which has been described in details elsewhere<sup>(7)</sup> consists mainly of a small Ludwig tube which feeds through

a supersonic nozzle a dielectric constant area channel ( $16 \times 3 \text{ cm}^2$ ) associated with a cold cathode electron gun and flush mounted discharge electrodes. The device is shown schematically on Fig.2.

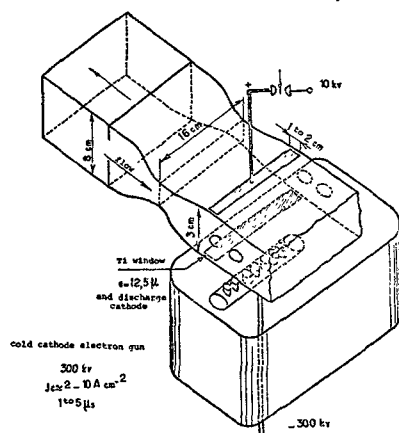


Fig.2 Schematic diagram of the facility.

The characteristics of the flow in the test section depends on the nozzle design, and two different types of nozzles have been used. Contoured nozzles permit to obtain a flow Mach number equal to 1.75, 2.5 or 3, the temperature being respectively equal to 120, 80 or 65 K and the density up to 2 amagats (rare gases  $\gamma = 1.67$ ). A screen nozzle has also been used permitting to achieve the following conditions in the test section :  $M = 2.0$ ,  $T \approx 100 \text{ K}$  ( $\gamma = 1.67$ ),  $\rho \approx 0.1 - 1 \text{ amagat}$ ). The electron gun energized by a Marx generator (300 KV - 1500 J) is fired when the flow is steady and excites the supersonically flowing gas mixture through a titanium foil. A non self-sustained discharge (glow) with an electric field up to  $E \approx 5 \text{ KV/cm}$  is added to the e-beam and applied between a 60 % transmission grid electrode near the e-gun foil and a facing flat electrode. The densities of transmitted electron beam and discharge current as respectively  $J_{e.b} = 3 \text{ A/cm}^2$  and  $J_D = 30 \text{ to } 200 \text{ A/cm}^2$ . A pair of gold mirrors (one with a 2 μm Hole) forms a resonant cavity integrated in the walls. This resonant cavity is set either at the discharge position or 3 cm downstream entirely outside of the discharge

in the last case, the time of flight between centers of discharge location and resonant cavity is equal to 20-30 μs. Experiments are also performed with this apparatus without flow, at room temperature with the same sample of gas mixture at the same density in order to obtain a direct comparison between low temperature flow and room temperature no flow laser characteristics.

The measured parameters consist mainly of :

- Marx generator, electron gun and flow time dependant characteristics.
- Time variation of laser intensity by means of 2 fast pyroelectric detectors with respectively 0.1 - 1000 μ and 7-1000 μ spectral response range. An infrared long pass filter,  $\lambda > 13.5 \mu$ , was set in front of one of the detectors. An home made infrared monochromator coupled with one of the pyroelectric detectors allowed to record laser power within a spectral band of 0.5 μ from 5 to 20 μ.
- Total extracted optical energy by means of joulemeters.

#### EXPERIMENTAL RESULTS

The described device has permitted to obtain the following experimental results :

- Stable electron beam sustained discharges have been achieved for various medium and high density laser mixtures in supersonic flow at very low temperature even for high loading conditions. In Fig.3 is shown a typical discharge current waveform obtained for an  $\text{Ar}/\text{CO}_2$  mixture in supersonic flow.

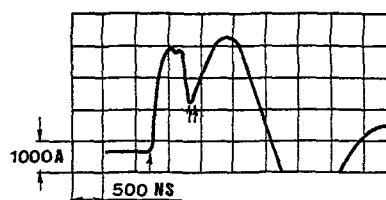


Fig.3 Discharge current time history for a typical e-beam sustained discharge experimental.  $\text{Ar}/\text{CO}_2$  (10/1),  $M = 1.75$ ,  $T \approx 120 \text{ K}$ ,  $V_c = 210 \text{ KV}$ ,  $V_D = 8 \text{ KV}$ .

High power short pulse laser emission at  $\lambda = 10.6\mu$  has been achieved when resonant cavity was set at the discharge position (gain switch mode) with Ar/CO<sub>2</sub>, Ar/CO<sub>2</sub>/H<sub>2</sub>, He/CO<sub>2</sub> and He/CO<sub>2</sub>/N<sub>2</sub> mixtures. For the conditions of these experiments lasing energy and power were several time higher at low temperature than at room temperature for the used mixtures and the same excitation and density. The time variation of laser power was very different at low temperature compared to the room temperature no flow case; in particular lasing appeared all the time earlier at low temperature and several very short laser spikes were observed for Ar/CO<sub>2</sub> and Ar/CO<sub>2</sub>/H<sub>2</sub> mixtures at low temperature indicating a strong modification in kinetics of laser medium. On Fig. 4, 5 and 6 are shown typical laser waveforms obtained with Ar/CO<sub>2</sub>, Ar/CO<sub>2</sub>/H<sub>2</sub> and He/CO<sub>2</sub>/N<sub>2</sub> mixtures respectively. Upper traces show room temperature no flow records while lower traces show low temperature records. It is noteworthy that the conditions of density, electron beam excitation and applied voltage are the same for all these experiments:  $\rho = 1$  amagat,  $j_{e.b.} = 3 \text{ A cm}^{-2}$ ,  $0.5\mu\text{s}$ ,  $V_{\text{capac.}} = 8.3 \text{ KV}$  and that attenuation and detector sensibility are the same for flow and no flow records.

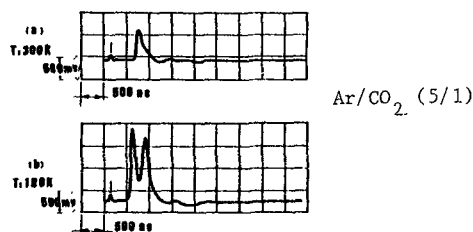


Fig.4 Typical CO<sub>2</sub> laser waveforms ( $\lambda = 10.6\mu$ ) for cavity center at discharge position.

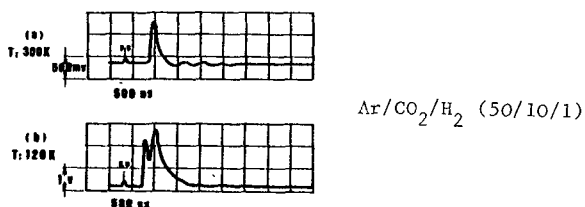


Fig.5 Typical CO<sub>2</sub> laser waveforms ( $\lambda = 10.6\mu$ ) for cavity center at discharge position.

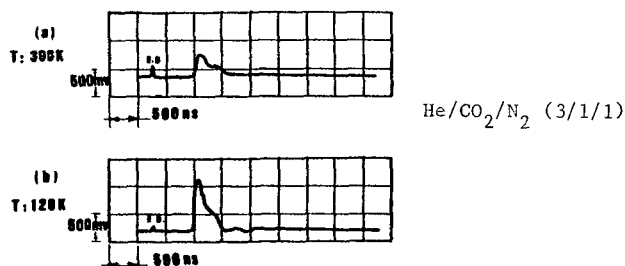


Fig.6 Typical CO<sub>2</sub> laser waveforms ( $\lambda = 10.6\mu$ ) for cavity center at discharge position.

- Laser emission several tens of microsecond long was achieved at  $\lambda = 10.6\mu$  when the resonant cavity was set 3 cm downstream of the discharge for He/CO<sub>2</sub>/N<sub>2</sub> mixtures in supersonic flow. The laser emission waveforms consisted of quasi-continuous power and several very short laser spikes as is shown on Fig. 7.

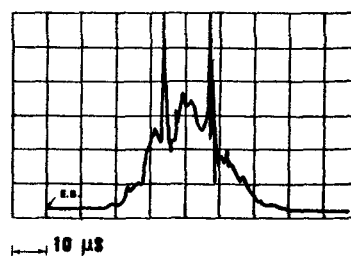


Fig.7 Typical CO<sub>2</sub> laser waveform for cavity shifted 3 cm downstream excitation zone. He/CO<sub>2</sub>/N<sub>2</sub> (3/1,1)  $\rho \approx 0.5$  amagat,  $M = 2$ ,  $T \approx 100^\circ\text{K}$ ,  $j_{e.b.} \approx 3 \text{ A cm}^{-2}$ ,  $V_{\text{capacitor}} = 9 \text{ KV}$ .

#### CONCLUSION AND FUTURE RESEARCH

Strong modifications of  $\lambda = 10.6\mu$  laser characteristics have been observed when various laser mixtures were cooled showing important modifications in kinetics of CO<sub>2</sub> laser.

Lasing at  $\lambda = 14\mu$ ,  $16\mu$  or  $18.4\mu$  wavelengths has not yet been achieved with the present device but these preliminary experiments seem to show that with Q switch techniques or with use of an external short pulse laser to saturate the  $10.6\mu$  transition it may be possible to populate the CO<sub>2</sub>

(100-020) vibrational levels during a short time (100-200 ns) and to obtain lasing on  $\text{CO}_2$  (100-010) and (02°0-010) transitions when a very high speed flow at low temperature and moderate density is excited by an e-beam sustained discharge either at the cavity position or slightly upstream. Possibility may also exist of short time inversion between (030) and (100)  $\text{CO}_2$  levels in  $\text{Ar}/\text{CO}_2$  mixtures following fast transfer between (030) and (010)  $\text{CO}_2$  vibrational levels.

Both experimental work at I.M.F.M. (gain measurement at  $10.6\mu$  by use of laser probe technique, Q Switching of laser emission) and numerical modeling of  $\text{CO}_2$  system kinetics at I.M.F.M. and Lebedev Institute of Physics will be pursued. If experimental demonstration of "cascade" lasing on  $\text{CO}_2$  can be made for I.M.F.M. conditions it would have important positive implications for development of high average power high repetition rate infrared laser sources useful for I.R. photochemistry.

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