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### **Optogalvanic Detection of CO<sub>2</sub> Laser Spectra in Electrodeless Radio Frequency Discharge Plasma**

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OPTOGALVANIC DETECTION OF CO<sub>2</sub> LASER SPECTRA IN  
ELECTRODELESS RADIO-FREQUENCY DISCHARGE PLASMA

Key Words : Infrared Absorption, Infrared Laser, Infrared  
Detection, Optogalvanic Effect, CO<sub>2</sub>, N<sub>2</sub>O, SF<sub>6</sub>, NH<sub>3</sub>

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ABSTRACT

CW line-tunable CO<sub>2</sub> laser-induced optogalvanic effects were observed from molecular plasmas excited by the electrodeless radio-frequency discharge. The optogalvanic effects were applied to detect laser output spectra and to optimize laser output power in CO<sub>2</sub> discharge plasmas and to measure optogalvanic(absorption) spectra in SF<sub>6</sub> and NH<sub>3</sub> plasmas. Other applications were briefly mentioned.

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### INTRODUCTION

The earlier experiments for  $\text{CO}_2$  laser-induced optogalvanic effect(OGE) were carried out in  $\text{CO}_2$  laser tubes or in external absorption tubes under the same experimental conditions as those in  $\text{CO}_2$  laser tubes<sup>(1)</sup>. The sizes of the absorption tubes were similar to those of laser tubes.  $\text{CO}_2$  and its mixtures were excited by DC discharges and irradiated with non-tunable(single-line)  $\text{CO}_2$  lasers. The OGEs in  $\text{CO}_2$  and mixture discharge plasmas have been applied to detect laser output power<sup>(2-4)</sup>, to align resonator automatically<sup>(3)</sup>, to stabilize laser frequencies<sup>(5-10)</sup>, and to measure vibrational relaxation times in  $\text{CO}_2$  molecules<sup>(11)</sup>.

It was already shown that by a similarity of the side-light spontaneous emissions between RF and DC discharges, physical processes in  $\text{CO}_2$  and  $\text{CO}_2\text{-N}_2$  mixture excited by RF discharge are the same as those by DC discharge<sup>(12)</sup>. It was also found that  $\text{CO}_2$  and mixture molecules in RF discharge produce more electrons and are less dissociated than those in DC discharge<sup>(13)</sup>. RF discharges, therefore, have been widely used for high-pressure  $\text{CO}_2$  waveguide lasers<sup>(14)</sup>.

In this letter,  $\text{CO}_2$  laser-induced optogalvanic effects were observed and applied to detect  $\text{CO}_2$  laser output spectra in  $\text{CO}_2$  plasmas and to measure optogalvanic(absorption)

spectra from  $\text{N}_2\text{O}$ ,  $\text{SF}_6$ , and  $\text{NH}_3$  molecular plasmas. Other applications were briefly mentioned. In these experiments, molecules in a small optogalvanic(discharge) tube were excited by an electrodeless RF discharge and irradiated with a CW line-tunable  $\text{CO}_2$  laser<sup>(15)</sup>.

#### EXPERIMENTAL

Fig. 1 is an experimental arrangement for optogalvanic measurements in molecular discharge plasmas. A conventional  $\text{CO}_2$  discharge laser(130 cm long and 2.0 cm in ID) provided a longitudinal mode which has a spacing of about 100 MHz and a bandwidth less than  $0.5 \text{ \AA}$ . Laser output power less than 1.0 W from a  $\text{CO}_2\text{-N}_2\text{-He}$  mixture was used to eliminate the saturation effects. The line-tunable  $\text{CO}_2$  laser from a vibrational transition of  $(00^0_1 - 10^0_0)$  was obtained by scanning a diffraction grating with 150 line/mm blazed at  $6.0 \text{ \mu m}$ . The single mode was selected by inserting an iris diaphragm into laser cavity. Laser beam was modulated by a mechanical chopper with a frequency of 500 Hz and irradiated through an optogalvanic tube. This tube was 20 cm in length and 1.0 cm in ID and had two salt(NaCl) Brewster windows.

Molecules were pumped out through the optogalvanic tube by a low-speed vacuum pump and pressure was monitored with a

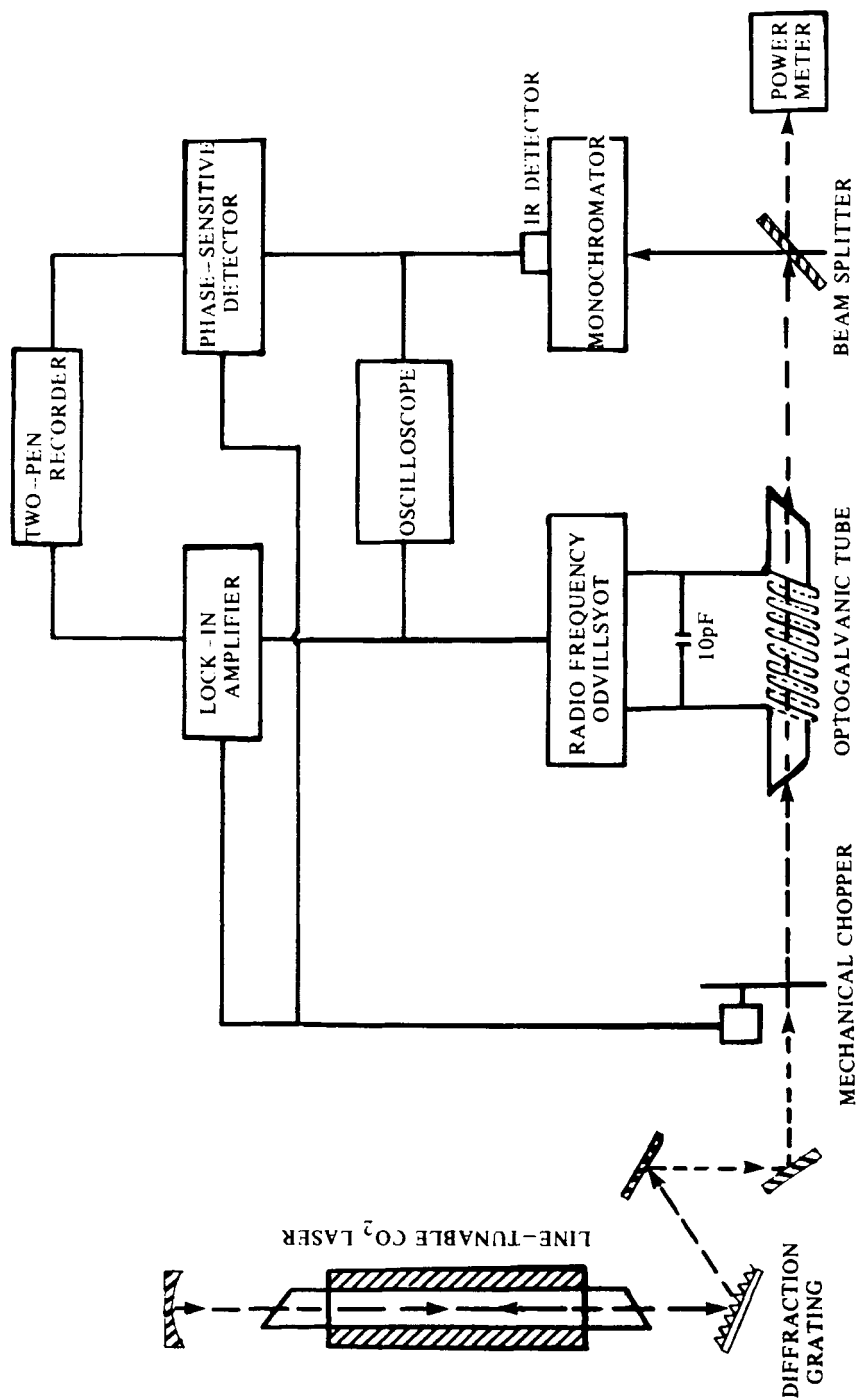


Fig. 1 Experimental arrangement for CO<sub>2</sub> laser-induced optogalvanic measurements

mercury manometer. In order to excite molecules, a Calpitts-type radio-frequency oscillator with a triode was used<sup>(16)</sup>. The RF oscillator consisted of a capacitor of 10 pF and a coil with 1.0  $\mu$ H(13 turns) and provided a frequency of 50 MHz. Molecules at 0.1 - 3.0 torr were easily discharged with input power less than 10 W.

Optogalvanic signals were detected by monitoring a change in voltage across a resistor of 7.8 K $\Omega$ . Laser beam transmitted through the optogalvanic tube was measured with a power meter or detected with a Hg-Cd-Tl photoconductor through a 1/4 m monochromator. Both optogalvanic and transmitted signals were sent into an oscilloscope or a Lock-in Amplifier and processed with integrating times less than 100 ms. The processed signals were recorded on a two-pen chart recorder.

#### RESULTS AND DISCUSSION

The electrodeless RF discharge not only excited easily low-pressure molecules but also provided quiet and stable (noise-free) plasmas. Therefore, the RF discharge provided two-orders of magnitude stronger OG signals than those in DC discharge under these experimental conditions.

By tuning wavelengths of CO<sub>2</sub> laser with a diffraction grating, the line-tunable laser radiation was selectively

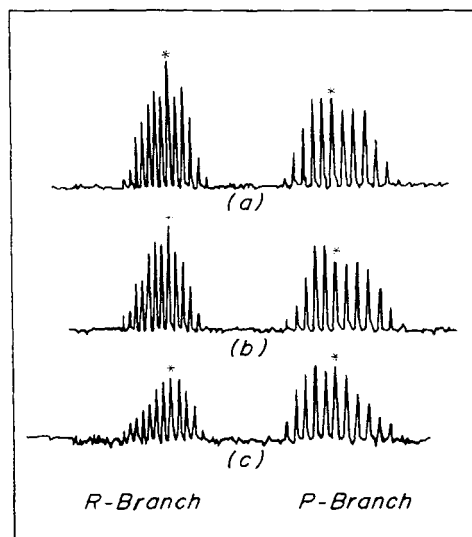


Fig. 2  $\text{CO}_2$  laser(a), transmitted(b), and optogalvanic(c) spectra from  $\text{CO}_2$  (0.5 torr) discharge. Spectra of (c) were magnified by a factor of 40. \*s are the corresponding lines, P(20) and R(20).

absorbed from molecular plasmas and provided OG signals, which are linearly proportional to laser output signals in the investigated laser power ranges.

Fig. 2 shows a comparison between optogalvanic and transmitted spectra from  $\text{CO}_2$  discharge plasma in a pressure of 0.5 torr and laser output spectra measured without gas and without discharge in optogalvanic tube.  $\text{CO}_2$  molecules were discharged with input power of 0.7 W. OG spectra were exactly same as laser output spectra. The OG signals were detected even very low pressures as long as discharge is

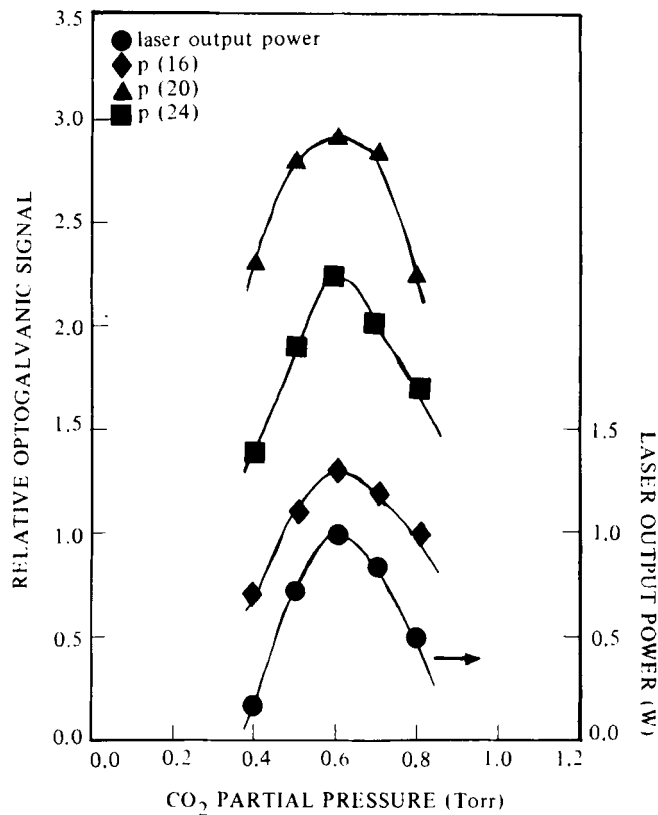


Fig. 3 Optogalvanic signals from CO<sub>2</sub> (0.5 torr) optogalvanic tube and laser output power as a function of CO<sub>2</sub> partial pressure in laser tube.

maintained. Such OG spectra in CO<sub>2</sub> discharge plasma were not obtained in DC discharge because of plasma noise.

Fig. 3 shows a strong dependence of OG signals upon laser output power irradiated. Laser output power was varied by adjusting CO<sub>2</sub> partial pressures of CO<sub>2</sub>-N<sub>2</sub>-He mixture in laser tube. OG signals in optogalvanic tube



increased with increasing laser power in laser tube. OGE in molecular plasmas, therefore, could be used to maximize or optimize laser output power in laser tube. It was also found that OG signals increase with increasing discharge input power and phases are reversed at higher input power. Negative OG signals at an input power of 1.0 W were reversed to positive at 7.0 W. This reverse in phase of OG signals was also observed at higher pressures, which is similar to that in DC discharge.

$\text{N}_2\text{O}$  laser spectra were also detected by optogalvanic effect. Several strong OG spectra were observed from  $\text{N}_2\text{O}$  discharge plasma in a pressure of 0.75 torr. These were obtained from lower rotational lines of R-branch transitions in  $10.8\text{ }\mu\text{m}$ - $\text{N}_2\text{O}$  laser and by higher rotational lines of P-branch transitions in  $10.6\text{ }\mu\text{m}$ - $\text{CO}_2$  laser. More detailed experiments of OGE in  $\text{N}_2\text{O}$  molecules are underway.

The OG spectra from  $\text{SF}_6$  discharge plasma were also observed (Fig. 4).  $\text{SF}_6$  molecules in pressure of 0.1 torr were excited with discharge input power of 5.0 W and irradiated with laser output power of 0.3 W. Strong OG signals were measured in strong absorption bands corresponding to hot bands of  $\text{SF}_6$  which are quasi-continuum near  $\text{CO}_2$  laser wavelengths of  $10.6\text{ }\mu\text{m}$ . More observation and discussion of OGE in  $\text{SF}_6$  molecules are found elsewhere<sup>(17)</sup>.

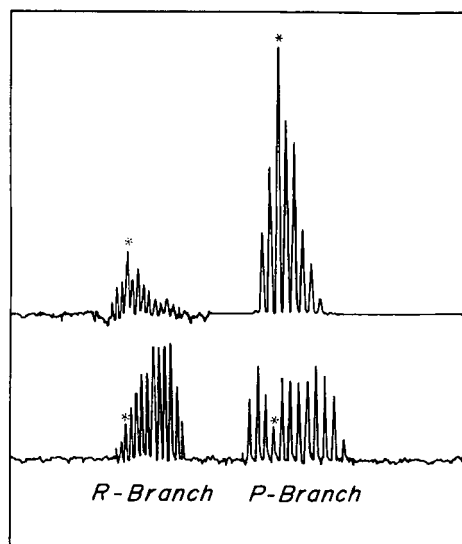


Fig. 4 Optogalvanic (upper) and transmitted (lower) spectra from  $\text{SF}_6$  (0.1 torr) discharge. OG spectra in R-branch were magnified by a factor of 10. \*s are the corresponding lines, P(16) and R(30).

From  $\text{NH}_3$  plasma excited with discharge input power of 16 W, several strong OG signals were also detected (Fig. 5). The OG signals from  $\text{NH}_3$  of a pressure of 2.0 torr were well consistent with strong absorption bands observed from a pressure of 760 torr by conventional absorption spectroscopy<sup>(18)</sup>.

#### CONCLUSION

It is believed that this is the first observation of optogalvanic spectra from  $\text{CO}_2$  discharge plasma which are

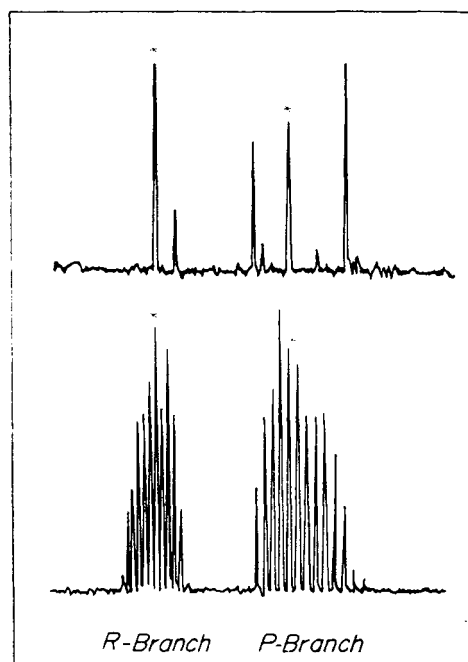


Fig. 5 Optogalvanic(upper) and transmitted(lower) spectra from  $\text{NH}_3$  (2.0 torr) discharge. \*s are the corresponding lines P(20) and R(20).

exactly same as  $\text{CO}_2$  laser output spectra.  $\text{CO}_2$  and other molecules were excited with an electrodeless RF discharge and irradiated by a CW line-tunable  $\text{CO}_2$  laser. This is a strong evidence of an interaction between vibrational populations in  $\text{CO}_2$  molecules and electron energy distributions by irradiation of laser beam ---- a change in populations between upper( $00^01$ ) and lower( $10^00$ ) vibrational states and/or ground state in  $\text{CO}_2$  molecules is resulted

from a change in gas temperature by a resonant absorption of laser radiation, thus affecting electron densities. These experiments also suggested that optogalvanic effects can be used to detect molecular spectra in discharge plasmas as an infrared detector which is inexpensive and fast-response.

By observing optogalvanic signals and absorption of molecules simultaneously in an optogalvanic tube, bandwidths in plasmas can be calibrated, thus measuring gas temperature. By controlling discharge current or voltage and then concentrations of ions in polyatomic molecular plasmas, optogalvanic effects can be applied to isotop separation. Tunable lasers in infrared regions make it possible to apply the optogalvanic effects to high resolution spectroscopy in molecules.

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