III. MOLECULAR ENERGY TRANSFER AND SPECTROSCOPY*

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RESEARCH OBJECTIVES AND SUMMARY OF RESEARCH

1. Resonance Fluorescence

On account of an optical mismatch of ~1.5 GHz between the 6328.17 Å line of the Ne laser and the nearest I_2 absorption line (see Quarterly Progress Report No. 86, page 9), the V' = 6 resonance fluorescence signal proves to be quite weak. Much stronger emission is observed from V' = 43 (Ar II 5145 Å line) and V' = 49 (Cd 5086 Å line); quenching measurements are being carried out on the latter state. In the oxygen system, the Hg line at 1849 Å lies closest to 8-0, R(11) in $^{16}O_2$ and the 8-0 band head in $^{16}O^{18}O$.[1]

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References

1. A. E. Douglas, Private communication, 1967.

2. Infrared-Optical Double Resonance

Saturation and Relaxation. A linear kinetic model for saturation of vibrational levels by intense infrared power has been developed and is now being numerically evaluated. Preliminary estimates indicate that in SF_6 at 0.5 Torr pressure, a significant fraction of the molecules may be pumped into an excited vibrational state when the gas is irradiated with ~100 W/cm^2 of 10.6-μ power. The predictions of the model are being tested by carrying out power absorption measurements in SF_6 and other systems.

CO_2 Laser Tuning by Selective Intracavity Absorption. See section III-A for a report on this research.

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(III. MOLECULAR ENERGY TRANSFER AND SPECTROSCOPY)

A. CO_2 LASER TUNING BY SELECTIVE INTRACAVITY ABSORPTION

Optical pumping with a CO_2 laser has already been used in an infrared-microwave double-resonance experiment[1] and is being contemplated for infrared-infrared and infrared-optical double-resonance experiments. In this connection, ability to tune across the P and R branches of the 00°1-10°0 transition greatly increases the number of coincidences with other molecular systems. Tuning should also enable one to pump different rotational levels in a given vibrational band.

With the optical pumping goal in mind, any tuning system employed should be as free from loss as possible. Otherwise the laser power, and consequently pumping efficiency, will be reduced. We observed that the method described here did not reduce the output power substantially. It also has the advantages of not requiring any great elongation of the cavity or the use of additional optical elements such as gratings or prisms. In other words, existing systems can easily be altered to utilize this tuning device.

The method consists of selective Q spoiling by introducing a known absorbing medium into the cavity, but separate from the discharge. See Fig. III-1.

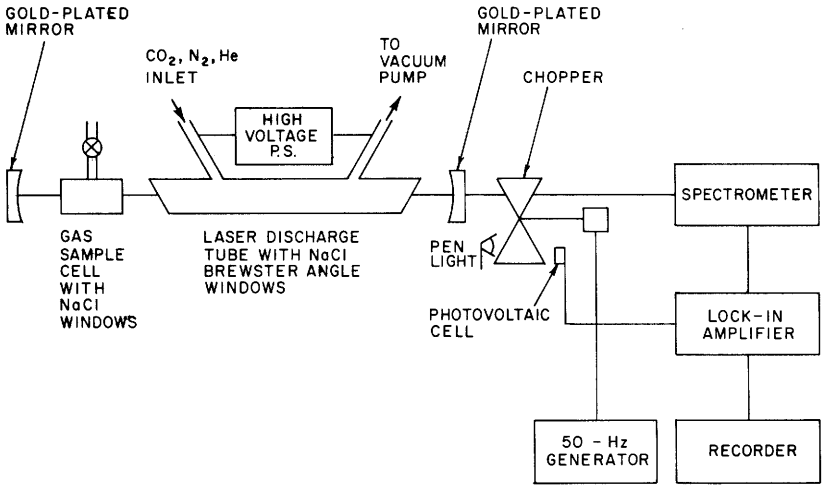

Fig. III-1. Laser apparatus.

Mixtures of SF_6 and He were used as the absorbing medium, but the method is obviously not restricted to this choice. The helium was used to decrease the lifetime of the ν_3 level of SF_6, thereby precluding any passive Q switching.[2] The results obtained with various gas mixtures are reported in Table III-1. All reported lines were cw operated with little or no power loss. Care was taken, however, to correct for longitudinal detuning resulting from the increase in optical length of the cavity when the cell was inserted.

Table III-1. Laser emission with SF_6 tuning.

Microns SF_6	Torr He	Active lines	Frequency (cm^{-1})
Evacuated Cell<10^{-5} Torr	Total	P(18), P(20)	945
3.67	.529	P(18)	946
6	1	P(20), P(22)	943
11.9	1.72	P(22) R(18), R(22)	942 976
13.6	4.2	P(20), P(22), P(24) R(22), R(24), R(26)	942 978
18	2.65	P(26) R(20)	939 976
25.6	8.1	R(18), R(20), R(22)	976
44.4	14	R(20), R(22)	977

The relation of the SF_6 absorption spectrum to the relevant portions of the emission spectrum of the CO_2 laser is given in Fig. III-2. The SF_6 absorption in the R-branch

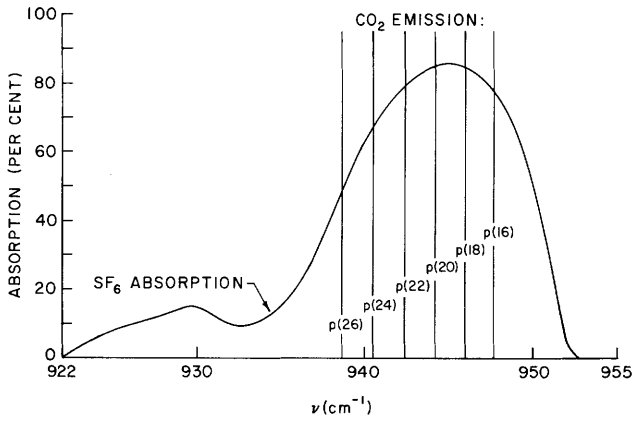

Fig. III-2. Prominent P-branch emission lines (arbitrary scale) and SF_6 absorption curve (P = 0.499 Torr in 10-cm cell).

region is less than 1%. The SF_6 spectrum was taken on a P. E. 521, at a gas pressure of 449 μ and with a 10-cm cell. The emission spectrum of the CO_2 laser is taken from Laures and Ziegler.[3]

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References

1. A. M. Ronn and D. R. Lide, Jr., J. Chem. Phys. 47, 3669 (1967).
2. O. R. Wood and S. E. Schwarz, Appl. Phys. Letters 11, 88 (1967).
3. P. Laures and X. Ziegler, J. Chim. Phys. 67, 100 (1967).