Notizen 1221

NOTIZEN

Investigations on the C2H2-O2 Chemical Laser

P. A. Büger, F. G. Sadie *, and O. G. Malan National Physical Research Laboratory C.S.I.R., P.O. Box 395, Pretoria, South Africa

(Z. Naturforsch. 28 a, 1221-1222 [1973]; received 21 April 1973)

Side light investigations of the flame in the laser cavity resulting from the oxidation of acetylene showed the presence of $\rm H_2O$ in this system. Stimulated emission in the fundamental band of CO is partially absorbed by the water formed in the reaction. This explains the very low total output of the $\rm C_2H_2-O_2$ chemical laser.

Continuous wave (cw) laser action resulting from the oxidation of acetylene has been reported ¹. The laser output was very sensitive both to changes in the total pressure and in the partial pressures of He, O₂ ann C₂H₂. This paper reports the results of investigations on this phenomenon and explanations are presented for the observed pressure dependence of the laser output.

The laser system has been described ^{1, 2}. Laser output both in the fundamental and overtone bands were analysed spectroscopically using a 1 m Czerny-Turner monochromator. The flame in the laser cavity resulting from the oxidation of acetylene was spectroscopically investigated side-on at two positions along the axis of the resonator (Figure 1). A small grating spectrometer was used for this purpose, the output of which was coupled through a photomultiplier, EMI type 9558, to a synchronous amplifier and recorder.

The following results were obtained:

- 1. Stimulated emission was observed in the fundamental band and in the overtone band of CO with a total output of about 30 μ W. The contributions of the individual bands were approximately equal.
- 2. The use of helium instead of argon as buffer gas increases the output only slightly in contrast to the marked increase observed in the outputs of both bands in the $CS_2 O_2$ chemical laser.
- 3. Side-light analysis of the flame showed the presence of the following band heads:

 $\begin{array}{lll} 3122\ \text{Å}\,(\text{OH}),\ 3872\ \text{Å}\,(\text{CH}),\ 4312\ \text{Å}\,(\text{CH}),\\ 4668\ \text{Å}\,(\text{C}_2),\ 5129\ \text{Å}\,(\text{C}_2),\ 6255\ \text{Å}\,(\text{H}_2\text{O})\\ \text{and}\ 6322\ \text{Å}\,(\text{H}_2\text{O})\,. \end{array}$

The variation in the intensities of these bands with discharge current 2 and their dependence on the partial pressures of He, O_2 and C_2H_2 are given in Figures 1 and 2.

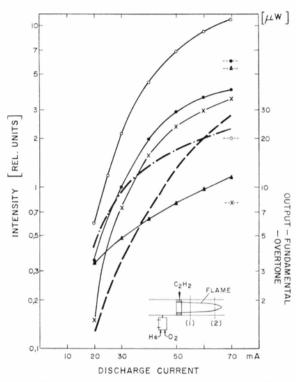


Fig. 1. Increase in intensity of distinct band heads with increasing discharge current. At 70 mA the intensities of the band heads measured at the tip of the flame are also given. The dependence of the output of both fundamental and over-

tone bands on the discharge current is shown. $-\circ - \mathrm{CH} - 4312\, \mathring{\mathrm{A}}, - \bullet - \mathrm{OH} - 3122\, \mathring{\mathrm{A}}, - \ast - \mathrm{C}_2 - 4668\, \mathring{\mathrm{A}}, - \triangle - \mathrm{H}_2\mathrm{O} - 6255\, \mathring{\mathrm{A}}, - \cdots - \mathrm{fundamental}, - - \mathrm{overtone}.$

4. The amount of water formed and collected in cold traps during reaction was larger than expected from the results of previous investigations ^{3, 4}. Water was detected spectroscopically, particularly at the tip of the flame.

The fact that fundamental band and overtone band outputs are approximately equal is most surprising. Normally the output of a fundamental band is approximately 1000 times higher than that of an overtone band. This was also demonstrated with the $CS_2 - O_2$ chemical laser 5, where the laser cavity, mirrors, window material, detecting and recording

^{*} On leave from the University of the Western Cape, Private Bag, Bellville, South Africa. Reprint requests to Prof. Dr. P. A. Büger, Optics and Spectroscopy Division, National Physical Research Laboratory, C.S.I.R., P.O. Box 395, Pretoria, South Africa.

1222 Notizen

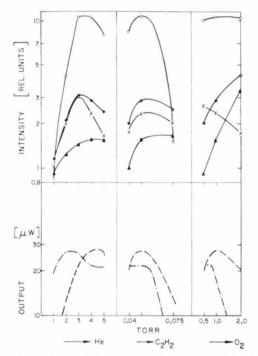


Fig. 2. Dependence of the intensity of distinct band heads on the partial pressures of He, C₂H₂ and O₂, and the output of fundamental and overtone bands as functions of the partial pressures.

$$-$$
0 − CH − 4312 Å, $-$ **•** − OH − 3122 Å, $-$ * − C₂ − 4668 Å, $-$ ∆ − H₂O − 6255 Å, $-$ · − fundamental, $-$ − overtone.

unit used were the same as in the present investigation. This effect could indicate the presence inside the laser cavity of an absorbent originating from the reaction between C_2H_2 and O_2 itself which absorbs fundamental band radiation but not radiation from overtone transitions. Our own measurements and published results 6 indicate that the most intense fundamental band transitions in the CO chemical laser occur around $1820\,\mathrm{cm}^{-1}$ and lower, in a range where H_2O lines 7 seriously overlap the CO-spectrum. Considering result 4 above and Figures 1 and 2, the assumption that water plays an important role as absorbent in the cavity is understandable and seems to be correct.

It is clear from Fig. 2 that an increase in the intensity of the $\mathrm{H}_2\mathrm{O}$ band head causes a strong decrease of the fundamental band intensity. The maximum output of the fundamental band is at

lower partial pressures than the maximum of the overtone band.

H₂O and OH bands at the tip of the flame become very intense with increasing discharge current. This may explain why the increase in the fundamental band intensity at low discharge currents is stronger than that of the overtone band and shows a slower increase with increasing current.

The most intense transition, considering the spectral output of the overtone band 1 , is at $4048 \ \rm cm^{-1}$ (transition 6-4) whereas the most intense transition in the CS_2-O_2 chemical laser is at $3728 \ \rm cm^{-1}$ (transition 12-10). The reaction mainly responsible for the production of vibrationally excited CO in the $C_2H_2-O_2$ chemical laser, i. e.

$$O + CH_2 \rightarrow CO + 2H$$
 (1)

has an enthalpy of $313.8 \, k \, \text{Joule mole}^{-1}$. In the case of the $CS_2 - O_2$ chemical laser the most important reaction is:

$$O + CS \rightarrow CO + S$$
 (2)

also with a heat of reaction of about 315 kJoule mole⁻¹.

The shift in the intensity distribution of the overtone spectrum thus cannot be explained by a difference between the exothermicities of these reactions. However, considering that the absorption of water increases sharply at about 4000 cm⁻¹, the shift in the intensity distribution can be explained.

Furthermore, the small enhancement of laser output using helium instead of argon as buffer gas in the C₂H₂ - O₂ system compared to the very strong increase in the output when argon is replaced by helium in the $CS_2 - O_2$ system, can also be explained by absorption of water. Helium reduces the rate of recombination of oxygen atoms created in the discharge, but enhances also the generation of water, thus only a small increase in output can be detected. The enhancement is also stronger in the overtone band than in the fundamental band where nearly no effect can be observed. Thus it is clear why stimulated emission in the $C_2H_2 - O_2$ chemical laser is very sensitive to changes in the total pressure. Also, the observation already mentioned 1 that the maximum total pressure at which gain still exceeds collisional deactivation is much lower than in the $CS_2 - O_2$ system is explained.

¹ F. G. Sadie, P. A. Büger, and O. G. Malan, Z. Naturforsch., in print.

² F. G. Sadie, P. A. Büger, and O. G. Malan, Z. Naturforsch. 27 a, 1260 [1972].

³ C. A. Amington, W. Brennen, G. P. Glass, J. V. Michael, and H. Niki, J. Chem. Phys. 43, 525 [1965].

⁴ R. Mavrodineanu and H. Boiteux, Flame Spectroscopy, John Wiley & Sons, New York 1965.

⁵ F. G. Sadie, P. A. Büger, and O. G. Malan, J. Appl. Phys. 43, 2906 [1972].

⁶ M. A. Pollack, Appl. Phys. Lett. 9, 237 [1966].

⁷ E. K. Plyler, W. S. Benedict, and S. Silverman, J. Chem. Phys. 20, 1178 [1952].