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Pulsed Chemical CO Laser
with Transverse Flow

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Abstract:

A chemical CO laser with transverse flow of premixed CS_2 and O_2 is described in which oxygen molecules are dissociated internally by a transverse pulsed discharge of up to loo Hz repetition frequency. The loo ground electrodes of this multispark discharge consist of 1 mm i.d. brass tubes serving simultaneously as gas inlet nozzles.

The maximum power during the $\sim$ 5o μsec long pulses is 6 watts at an operating pressure of $\sim$ 7o torr and gas flow rates of $CS_2 : O_2$ $\sim$ lo^{-5} :lo^{-3} moles/sec. The spectrum consists of about 5o P branch lines with upper vibrational quantum numbers 3 - 12 and rotational quantum numbers 15 - 2o.

The efficiency of this laser of η = o.o5 % is slightly higher than that of a commercial He Ne laser, but falls short of the high values which can be achieved with electrically pumped CO lasers.

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Introduction

All chemical laser systems have to start from stable molecules such as H_2, SF_6, O_2, which react after at least one of the partners has been dissociated initially. The chemical reaction then leads to a vibrationally excited molecule, the active laser substance. The initiating dissociation can be obtained by photolysis /1/, /11/ by heating the gas /2/ or by electron impact in low pressure electrical discharges /3/, /4/, /5/.

One of the known systems is the chemical CO laser based on the reactions /6/

$$O + CS_2 \longrightarrow CS + SO$$
$$O + CS \longrightarrow CO^+ + S$$
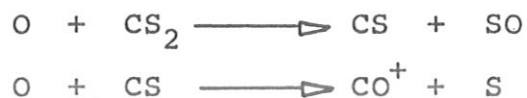
Oxygen atoms are required to initiate this reaction.

CO is an interesting molecule, because it has better storage capacity than the HF molecule (longer vibrational relaxation time τ_{vv}) it "lases" at a convenient intermediate wavelength range $\lambda \approx 5$ μ, it is easier to handle than for instance HF, and it can be excited not only by chemical reaction /3/, /4/, /5/ but also by gasdynamical expansion /7/ in Laval nozzles and by direct electron impact /8/, /9/.

This report deals with an experiment which was set up firstly to learn about CO as a lasing substance, and secondly to study one particular laser initiation method, namely the dissociation in an electrical discharge. (Photolytical and thermal dissociation are applied in two concurrent experiments, ASTERIX and SCHAUDER, in our laboratory.)

The intention was further to combine the known construction principles of Beaulieu's /1o/ TEA laser and a transverse flow system, hopefully to obtain higher laser powers than reached in previously reported experiments. Trying a transverse flow system with CO molecules in the TEA laser tube also led to the name of the experiment: TEA & CO.

Historically the first chemical CO laser of Pollack (1966) /1/ was initiated with a flash lamp and produced vibrationally exited CO molecules by the chemical reaction of oxygen with carbon disulfide. The laser produced 10^{-5} Joule per pulse with an average power of o.5 W. The total efficiency of converting the 2ooo Joule of electrical energy stored on the condensor bank into laser light was only $\eta_{el} =$ 5.10^{-9}. The first continous chemical CO laser reported by Suart, Kimbell and Arnold (197o) /3/ had a power of a few mW and utilized a flow system parallel to the resonator axis. (similar experiments were carried out simultaneously by Wittig et al. /4/.) The oxygen atoms were generated outside the laser cavity in a microwave discharge. An increase of the laser power of about a factor lo was reached in a transverse flow system by Jeffers and Wiswall (197o) /5/. Again the oxygen atoms were generated outside the laser cavity prior to mixing the two gas components. Wall- and volume recombination of oxygen atoms during the transit time of the oxygen from the dissociation area into the reaction area is unavoidable in these experiments, although it is less pronounced in Jeffers design.

In order to increase the power of a chemical CO laser even further, we have practically eliminated the wall recombination losses by dissociating the oxygen directly in the laser cavity in a transverse (necessarily pulsed) discharge. A transverse flow through the loo tube shaped electrodes guarantees a rapid exchange of the reaction products. The average power during each pulse is 1 watt. The overall efficiency:laser energy/electrical energy is $\eta_{el} = 5.10^{-4}$, a value which is about equal to the efficiency of a commercial HeNe laser. It is however very small compared to the efficiency $\eta_{el} = 0.12 \rightarrow 0.17$ of the electrically excited CO laser of Bhaumik (197o) /9/, and we will show, that the chemical CO laser can in principle not reach the high efficiency of the electrically excited laser.

Design of the Laser

A schematic view of the laser reaction chamber is shown in Fig.1. Oxygen and carbon disulfide enter separated or premixed through 2oo small tubes (1 mm i.d.) which are arranged in two rows and connected to two brass supply tubes A, B. One set of tubes is made of brass and serves as ground electrodes the other set consists of glass tublets. The depths h_1 and h_2 can be varied.
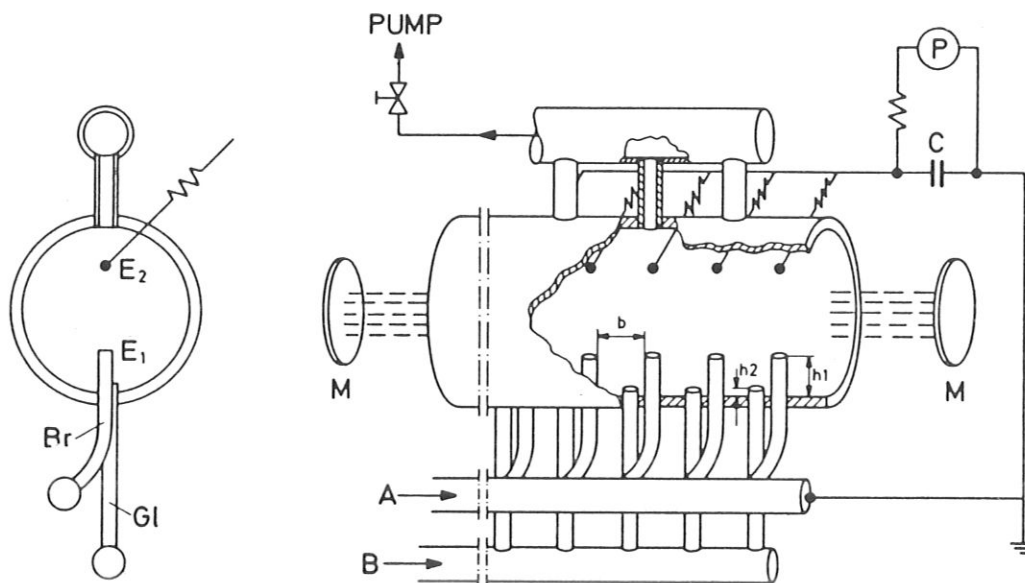
Fig.1   Laser reaction tube. Br = brass tube, Gl = glass tube,
M = laser mirror, P = power supply 6o mA, D = 3o kV,
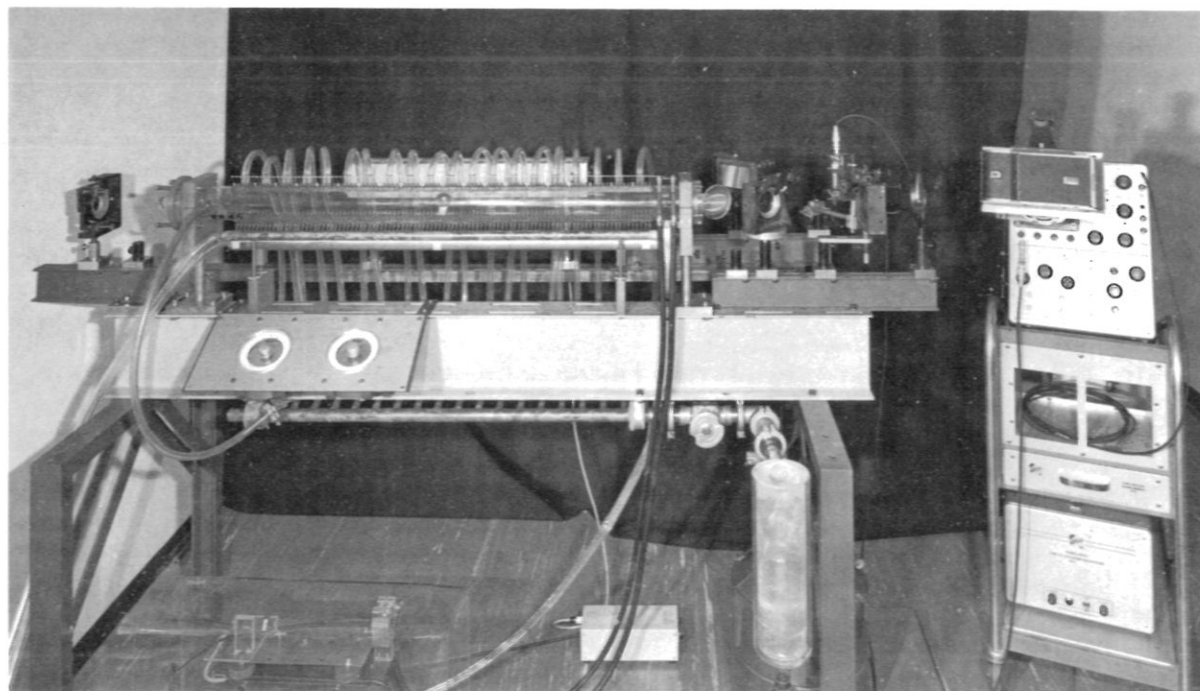C = 12 nF condensor, E_1, E_2 = electrodes

Fig.2 "TEA & CO" chemical CO laser

In order to guarantee homogeneous dissociation in the reaction volume of any desired length and relative high and variable pressure, a transverse multi spark discharge similar to the TEA design of Beaulieu /10/ was chosen. The legs of the 100 resistors, R = 1 kΩ, are mounted opposite to the gas inlets and serve as high potential (negative) electrodes. The resistors are connected in parallel via a trigger spark gap to a 12 nF condensor, charged by a 0 - 30 kV, 60 mA power supply P. The sparks have a decay time of typically 1 μsec in the pressure range 10 - 100 torr. Pulse repetition rates up to 100 sec^{-1} are possible. From the decay time of the electric pulse and the known external resistance an internal resistance of 8 kΩ is calculated for each of the h_3 = 3,0 cm long sparks for a charging voltage of 11 kV, Each spark carries a maximum current of 1,6 Amp through the visually observed cross section of about 0.2 cm^2.

The burned gas is extracted by a mechanical pump through 20 tubes (1 cm i.d.). The valve in the pump line and the flow meters in the supply line of each chemical constituent facilitate the independent adjustment of pressure, composition and gas flow rates. The 7 cm i.d. lucite reaction tube is 120 cm long (20 cm longer than the rows of gas inlet tubes) and it is sealed with K Cl windows mounted at the Brewster angle. The purpose of having one set of insulating gas inlet tubes was to aim the dissociation mainly at the gas flow through the electrode - tubes. No difference however was found in the laser emission when passing the oxygen through one or the other set. For that reason the gases were later premixed in most of the experiments, using the brass electrodes only. Oxygen of 99.9 % purity was taken from a cylinder, CS_2 gas was obtained by evaporating liquid CS_2 at room temperature. A full view of the laser is given in Fig. 2.

The laser cavity is 180 cm long and has one R = 5 m gold coated mirror on one end and a coated germanium plate of 40 % transmission on the other side. Some experiments were carried out with a flat gold mirror and a KCl plate for power extraction replacing the germanium plate. For relative measurements a Ge:Au detector at 77 ^{o}K (Ford Philco CPC 215) or an uncooled InSb detector (Valvo ORP 10) was used. With the high repetition rate of the laser it was however easy to calibrate these detectors against a standardized cone calorimeter (CDC Control Data Model 101 Thermopile). The absolute energy measurements compared well with an energy estimate based on the colour change of a liquid crystal /12/.

Experimental results

To test the TEA & CO laser cavity and the detector the laser was operated as an ordinary CO_2 laser using a mixture of CO_2 and N_2 and He and filling pressures in the range 5 - 4oo torr. These CO_2 pulses have the familiar peak with short rise time, about 1 - 2 μsec after initiation of the discharge. The laser was then operated as a chemical laser with CS_2-O_2 mixtures. CO-laser emission was observed in the range of filling pressure $3 \leq p_t \leq 15o$ torr. Gas flow rates of $CS_2 : O_2 \approx 1o^{-5} : 1o^{-3}$ mole/sec gave good results. A typical laser pulse is shown in Fig.3. To measure the spectrum the laser beam was collimated with a 9,7 cm focal length KCl lense onto the entrance slit of a 1m-McPherson monochromator with a 15o lines/mm grating. About 5o rotational vibrational P branch lines were found in the range 49ooo - 56ooo $\overset{o}{A}$, with vibrational quantum numbers of the upper state 3 - 12 and rotational quantum numbers 14 - 2o, see Appendix.
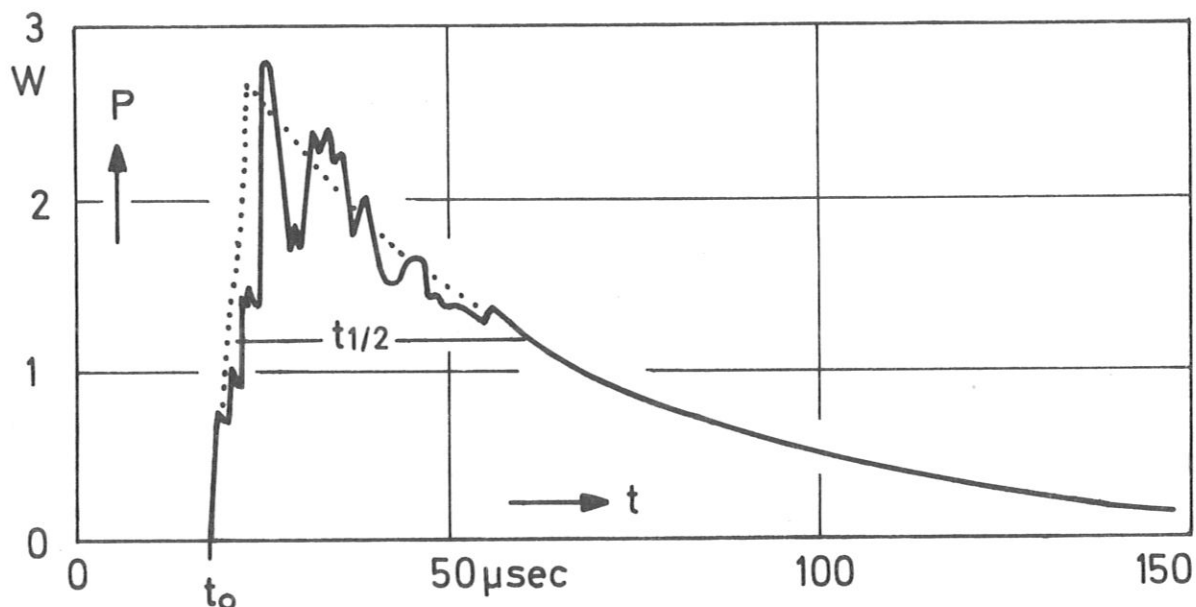

Fig.3   Typical laser pulse obtained with $p_t = 2o$ torr, $CS_2:O_2 = 1.2 \cdot 1o^{-5}:2.1o^{-3}$. Charging voltage 1o.5 kV.

The laser pulse is delayed by $t_o \approx 1o \rightarrow 2o$ μsec with respect to the electrical pulse. The risetime of about 1 μsec is independent of the filling pressure. Laser emission is confined to an area of about 5 x 15 mm centered on the axis of the reaction tube, which is about 25 mm away from the ground electrode. The oxygen is apparently produced in this active area, as desired originally, since the delay

time t_o is not long enough for atoms from other parts of the reaction tube to diffundate or flow into this area.

Laser action with the same delay time t_o is observed in fresh mixtures as well as in partly burned mixtures. Therefore electronic excitation of CO molecules in the discharge can be excluded, and we have a laser with chemical pumping, as wanted.

The average laser power during the pulse of about 1 W is considerably higher than the power of chemical CO lasers with external oxygen generation /3/, /5/. However in order to extract this power continously one would need a power supply capable of charging the condensor about 10^4 times per second. (Under DC conditions it would not be possible to operate the 100 parallel discharges from a single supply.) With our charging unit, pulse rates of 100 sec^{-1} were possible (0.01 duty cycle), so that the power averaged over a full charging period was p = 10 mW. The laser power increases linearly with the energy on the condensor. The energy per pulse is about 0.1 mJoule.

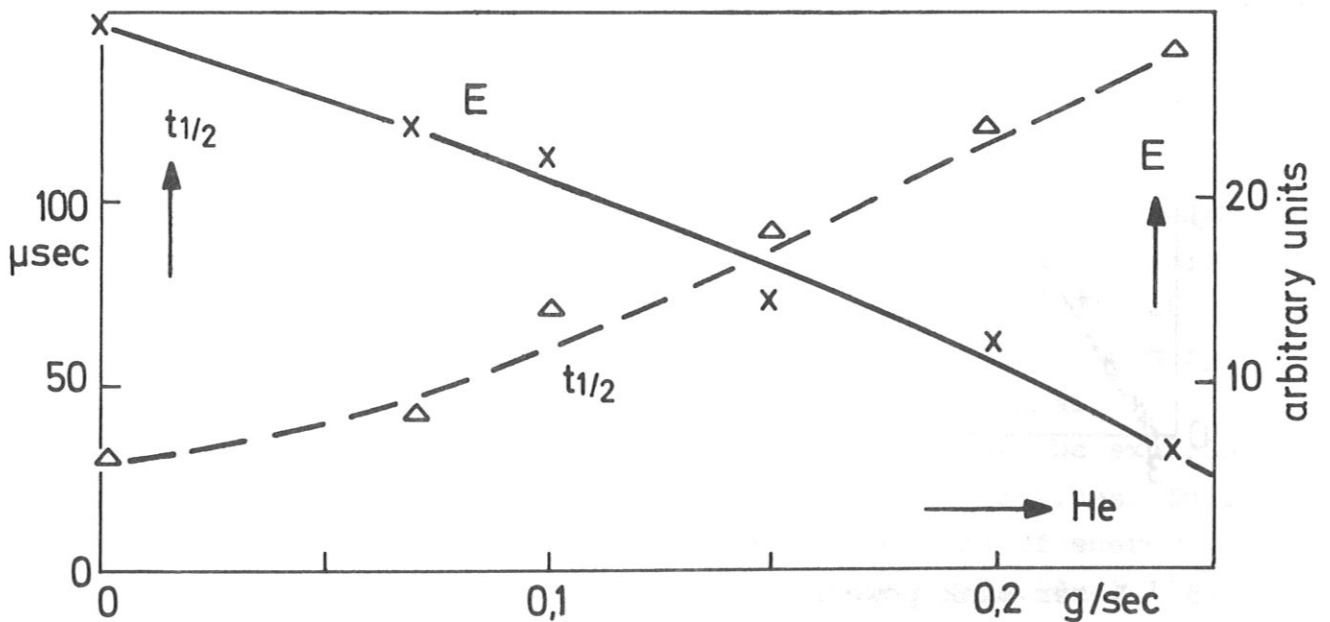Fig. 4   Energy E and decay time $t_{1/2}$ of laser pulses for various amounts of He added. p_t = 30 torr, $CS_2 : O_2 = 1.2 \cdot 10^{-5}$: $1.7 \cdot 10^{-3}$ moles/sec charging voltage 9 kV

Addition of helium at constant pressure and constant CS_2 and O_2 flow rates increases the decay time of the pulse, but reduces the total laser energy, see Fig.4. A similar reduction of laser energy is observed if nitrogen is added to the flow.

If the pressure p_t is increased in the reaction tube while the flow rates are kept constant the laser power first rises to reach a maximum at $p_t \approx 7o$ torr and drops at higher pressures to be hardly measurable at $p_t = 15o$ torr. Fig.5. The pulses are very reproducable at low pressures but fluctuate by about 25 % in the pressure range of highest laser emission.
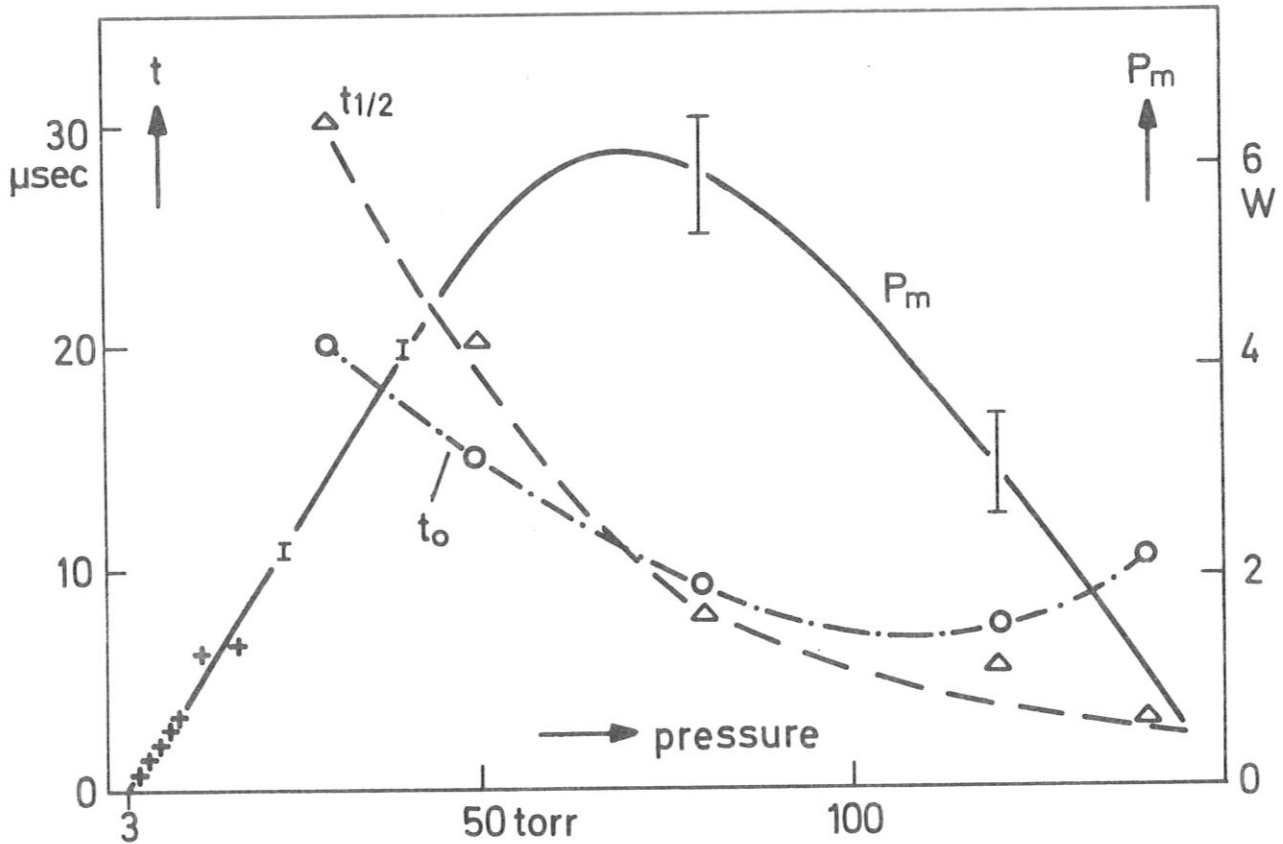

Fig.5   Laser peak power P_m of CO laser as function of total filling pressure p_t. $CS_2:O_2 = 1.2 \cdot 1o^{-5}:6 \cdot 1o^{-3}$ moles/sec.

The initial increase of laser power with pressure will be discussed below. The decrease at higher pressures has probably three reasons. Firstly the gas temperature, which is a few hundred degrees

in an electrical discharge of p_t = 1 torr, Hoyaux (1968) /13/, rises
by about a factor 1o if the pressure is increased to p_t = 1oo torr.
The laser lines are therefore homogeneously broadened by an increase
of the Doppler width as well as by pure pressure broadening. This
homogeneous line broadening increases the threshold for inversion
and thereby reduces the extractable laser power. Secondly the in-
version itself is reduced by deactivating collisions at higher pressures.
The vib-vib relaxation times for instance go drastically down. According
to Yardley (197o) /14/ $p_{CO} \cdot \tau_{vv}$ = 2o8 /usec torr, i.e. the relaxation
time τ_{vv} decreases by a factor 1o if the pressure is raised from 15
to 15o torr. Thirdly the electron temperature falls if the pressure
is raised, Fig.6, so that the electrons are less effective in
dissociating the oxygen molecules necessary for the reaction.

Efficiency

The electrical power to operate the laser was obtained from a
current and voltage measurement(at the power supply) with the laser
operating at 1oo pulses per second. The electrical energy per pulse
could also be obtained from the charging voltage and the capacity
of the condensor. The efficiency of converting the electrical energy
could hence be determined. For single pulse operation as well as for
pulses at high repetition rates an efficiency

$$\eta_{el} = 0.05 \%$$

was found. This value is comparable with the efficiency of a commercial
He-Ne laser and lies well above the efficiency of the first chemical
CO laser of Pollack with η_{el} = $5.1o^{-9}$. However our result is still
very low compared with the efficiency η_{el} = 12 - 17 % obtained by
Bhaumik /9/ in an electrically exited CO laser with special
additives (Xe and Hg). This difference in efficiency may be explained
firstly by the fact that Bhaumik observes only 1o laser lines. In our
laser, emitting about 5o lines, a much larger fraction of energy is
lost below the threshold of laser emission of each individual line.
Secondly it is more efficient to excite the vibrational states of a
CO molecule directly by electron impact than indirectly via a chemical
reaction. This second point is now explained in more detail.

The most likely pumping mechanism of the CO laser /6/ is

$$O + CS_2 \longrightarrow CS + SO, \quad O + CS \longrightarrow CO^+ + S$$

One O_2 molecule therefore has to be dissociated in order to generate one vibrationally exited CO molecule, which may issue β_{ch} vibrational quanta at a wavelength of about 5 μm. Dissociation in an electrical discharge occurs by electron impact, since firstly energy transfer is more likely between electrons and heavy particles and secondly the electrons are much "hotter" than the atoms and molecules in low pressure electrical discharges, see Fig.6.
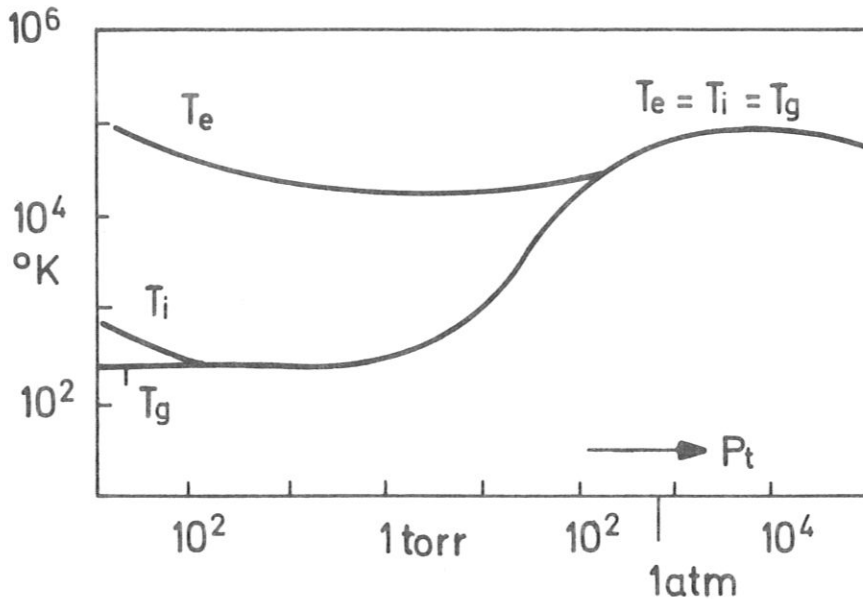

Fig.6   Gas and electron temperature at high and low pressures, according to Hoyaux 1968 /13/.

The number of available quanta per unit volume can be approximated as

$$n_q = \beta_{ch} n_{CO^+} \leq \frac{1}{2}\beta_{ch} n_O$$

where the number density of oxygen atoms n_O, obtained by electron impact dissociation of oxygen molecules, can be calculated from a collision integral.

$$\frac{dn_o}{dt} = 2 \int\limits_0^\infty\int \widetilde{\sigma_o}(v_e)\, n_{o_2} f(v_{o_2})\, n_e\, f(v_e)\, v_e\, dv_e\, dv_{o_2} \tag{2}$$

Here we have neglected the velocity of the oxygen molecules v_{o_2} compared to the velocity v_e of the electrons. The first integration over dv_{o_2} can be carried out immediately. Since $f(v)dv = :1$, we have

$$\frac{dn_o}{dt} = 2\, n_{o_2} n_e \int\limits_0^\infty \widetilde{\sigma_o}(v_e) \cdot v_e \cdot f(v_e)\, dv_e = 2\, n_e\, n_{o_2}\, k_o(T_e) \tag{3}$$

The integral can be evaluated if the velocity distribution of the electrons and the dissociation cross section $\widetilde{\sigma}_o(v_e)$ is known. The latter was measured by Craggs et al. /16/. His values are displayed in Fig.7. A Maxwell velocity distribution was assumed for the electrons, with temperatures which are typical of low pressure discharges
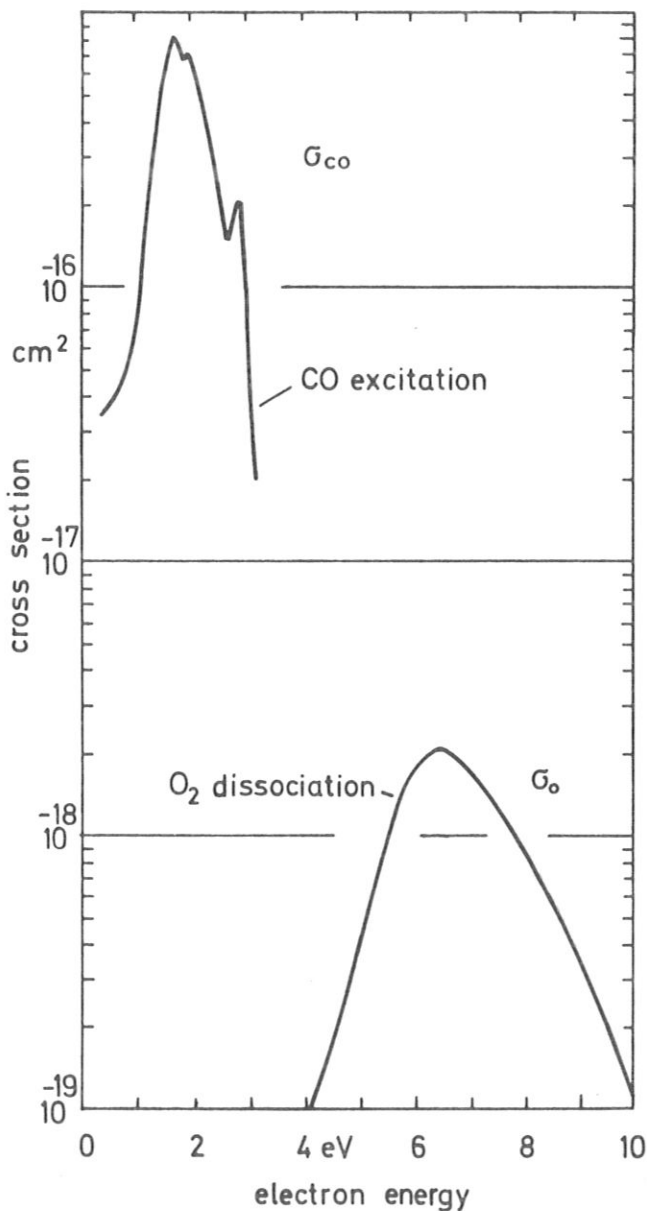

Fig.7   Collision cross section for CO excitation /16/ and oxygen dissociation /15/ by electron impact.

The integration was carried out numerically for three representative temperatures, Table I

T_e	$k_O(T_e)$ cm^3/sec	$k_{CO}(T_e)$ cm^3/sec	$\eta_{el}/\eta_{chem}=\beta_{el}k_{CO}/\beta_{ch}k_O$ $= F$
$10,000\ ^oK$	$2.8 \cdot 10^{-12}$	$1.5 \cdot 10^{-8}$	$1.1 \cdot 10^3$
$20,000\ ^oK$	$2.5 \cdot 10^{-11}$	$1.6 \cdot 10^{-8}$	$1.3 \cdot 10^2$
$30,000\ ^oK$	$4.1 \cdot 10^{-11}$	$1.2 \cdot 10^{-8}$	$0.6 \cdot 10^2$

Table I   Collision integrals $k_O(T_e)$ for generation of oxygen atoms
and for the excitation $k_{CO}(T_e)$ of CO molecules. $\beta_{ch} = 10$,
$\beta_{el} = 2$ assumed.

With the collision integral one can obtain the number of oxygen atoms produced under typical conditions of the discharge in our TEA & CO laser.

$$\Delta n_O = \Delta t\ 2n_{O_2} \cdot n_e \cdot k_O(T_e) \geqslant 2\Delta n_{CO}^+ \tag{4}$$

With $n_{O_2} = 10^{17}\ cm^{-3}$, an estimated 0.1 % ionization,
(so that $n_e = 10^{14} cm^{-3}$) and an electron temperature $T_e = 20,000\ ^oK$
one obtains $\Delta n_O = 5.10^{14}$ within a time interval of $\Delta t = 1\ \mu sec$.
Estimating the lasing volume of the 100 parallel sparks in the
TEA & CO laser to be 50 cm^3, a total of $N_O = 10^{16}$ oxygen atoms
would be produced, compatible with the 3.10^{15} laser quanta measured
externally. According to equation (4) the oxygen production rate
is linearly proportional to the initial oxygen density n_{O_2}. We have
observed the related increase of laser power with initial pressure
p_t, Fig.5. At pressures $p_t \approx 50$ torr and higher the laser emission
is inhibited by other effects (see above).

The rate of producing oxygen atoms in the electrical discharge
must be compared with the rate of producing vibrationally excited
CO molecules in a comparable discharge. The rate is obtained from
a similar collision integral

$$\frac{dn_{CO^+}}{dt} = n_{CO}n_e \int_0^\infty \tilde{\sigma}_{CO}(v_e) \cdot v_e \cdot f(v_e) \, dv_e = n_{CO} \cdot n_e \cdot k_{CO}(T_e) \tag{5}$$

where $\tilde{\sigma}_{CO}(v_e)$ is the collisional excitation cross section of CO molecules for vibrational excitation. This overall cross section for **excitation** of vibrational quantum numbers $v = 1$ to $v = 8$ was measured by Schultz (1964) /17/, see Fig.7. Taking his data, numerical values for the collision integral $k_{CO}(T_e)$ were calculated and listed in Table 1. On the average $\beta_{el} \approx 2$ vibrational quanta are **excited** by electron impact. (The energy at the maximum of $\tilde{\sigma}_{CO}$ corresponds to 7 vibrational quanta.)

The large difference of the integrals $k_O(T_e)$ and $k_{CO}(T_e)$ is due to the fact, that the dissociation cross section for O_2 is about a factor 2o smaller than the exitation cross section of CO, and furthermore $\tilde{\sigma}_{CO}$ peaks near the maximum of a $T_e = 2o,ooo$ ^oK Boltzmann distribution of the electrons, wheras σ_O has its maximum at about 7 eV, where the velocity distribution of the electrons has already dropped to very low values.

The collisional integrals k_O and k_{CO} may be used to estimate the efficiency of pumping energy into vibrational excitation of the CO molecule. During the interval Δt a number of

$$\Delta q \, (el) = \beta_{el} \, n_e \, n_{CO} \, k_{CO}(T_e) \, \Delta t \tag{6}$$

vibrational quanta are obtained by collisions between electrons and CO molecules, where β_{el} is the average number of vibrational quanta per electron impact. Schulz measured excitation up to the level $v = 8$. From his collision cross sections for the individual vibrational levels the rate coefficients $k(v = 1)$ to $k(v = 8)$ were calculated for $T_e = 2o,ooo$ ^oK, and an average excitation per collision $\beta_{el} = 2$ was found.

In comparison to (6), chemical pumping via oxygen dissociation yields at best

$$\Delta q \, (chem) = \beta_{ch} \frac{1}{2} \, n_{CO}$$

vibrational quanta, where $\beta_{ch} = 1o$ is the average vibrational excitation immediately after the chemical formation of the CO molecule, and n_{CO} is given by (4).

The ratio (see Table I)

$$F = \Delta q(el)/\Delta q(chem) \approx 2k_{CO}/10k_O \approx 10^2$$

expresses how many more vibrational quanta of the CO molecules are produced directly by electron impact than by chemical reaction of CS_2 and oxygen atoms, generated in the first place by electron collisions. The numerical value of F is the upper limit by which the efficiency of the electrically excited CO laser may exceed the chemical CO laser.

While the excitation by electron impact is very effective, the mechanism of establishing inversion among the vibrational states may take up much or even nullify the advantage of the electrically excited CO laser. The molecules in the chemical laser have a complete inversion and high excitation immediately after the reaction. In the electrically excited CO laser the inversion must be generated and collisions are required which depopulate the lower vibrational states. The energy may be transferred to certain additives, such as Xe and Hg /9/ or to the ground state /8/ according to the scheme

$$CO(v=0) + CO(v) \longrightarrow CO(v=1) + CO(v-1)$$

Due to the unharmonicity of the CO molecule this energy transfer is effective only up to a level v_e where the kinetic energy of the colliding molecules suffices to make up the energy difference of the two quanta $v \rightarrow v-1$ and $v=0 \rightarrow v=1$. Graham calculates v_e as function of the gas temperature:

$$v_e = 1 + kT/2hc\omega_e x_e$$

where k, h, c are taken in cgs units, T in $^\circ K$ and $\omega_e x_e$ has the value 13,461 cm^{-1} for the CO molecule.

In order to utilize much of the vibrational excitation of the CO molecule for laser emission, v_e must be kept low, which requires the gas temperature to be small. For CO gas of room temperature a value $v_e = 9.6$ is found. Hence all excitation energy in levels below $v=9$ is wasted and cannot contribute to laser emission. The rates of exciting levels above $v=9$ by electron impact are about two orders of magnitude

smaller than the total rate coefficient k_{CO} listed in Table I /19/ so that electrical excitation ceases to be a competition for the chemical excitation. At gas temperatures of a few hundred degrees which can be expected in a reacting mixture of oxygen and CS_2 as well as in gas discharges in p>1o torr practically all vibrational levels can exchange energy with the ground state so that the inversion of a chemical CO laser is quickly reduced and electron impact excitation will not lead to an inversion at all.

Conclusions

The TEA & CO laser with transverse flow and transverse multi spark discharge for the initiation of the reaction of oxygen and CS_2 reaches pulse powers of up to 6 watts, which is about two orders of magnitude above previous chemical CO lasers /5/. With pulse durations of $\sim$ 5o /usec and pulse repetition frequencies of 1oo Hz the laser operates like a CW light source for many practical applications. The power has a maximum at an operating pressure of $\sim$ 7o torr. About 5o lines in the range 49ooo to 56ooo A are emitted simultaneously. The efficiency of the laser of η = o.o5 % could possibly be improved by the use of other oxydizing molecules (such as O_3, NO_2) with lower dissociation energy than O_2. The laser reaction vessel itself is quite versatile: it has also been operated as a standard TEA laser with CO_2 gas and with pure oxygen gas. The first medium produced the well known short spikes, of typical 1 /usec length, the second gas yielded much longer pulses of about 5oo /usec duration.

Appendix

The spectrum of the CO laser was measured with a 1m-McPherson monochromator, calibrated against strong lines of Hg, Na, Cd and the red HeNe laser line. High orders (8th to 12th) had to be used in order to cover the spectral range of 48ooo to 58ooo A, so that the wavelength corrections for air (taken from Handbook of Chemistry and Physics) had to be applied. The spectrum was compared with vibrational rotational transitions of the CO molecule calculated by Proch and Wanner /18/. The measured wavelength and the tentative identifications are given in Table II.

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      cross sections. They fall on a straight line with lope - 2 when
      plotted   versus v on a log-log diagram, extrapolation to levels v 8
      is hence possible.

Table II

Spectrum of the CO laser operated at: p = 25 torr,
$CS_2:O_2 = 1 \cdot 10^{-5} : 6 \cdot 10^{-3}$ moles/sec, 11 KV, Pulse repetition rate
about 25 Hz.

Wave length	identification V_{upper}		Wave length	identification V_{upper}	
49152	3	P(14)	52444	7	P(2o)?
49254	3	P(15)	52600	8	P(15)?
49362	3	P(16)	52716	8	P(16)?
49454	3	P(17)	52822	8	P(17)
49656		?	52936	8	P(18)
49760	4	P(14)?	53046	8	P(19)
49894	4	P(15)	53160	8	P(2o)
49994	4	P(16)	53218		?
5o2oo	4	P(18)?	53274		?
5o548	5	P(15)	53328		?
5o654	5	P(16)	53676	1o	P(12)?
5o744	5	P(17)	5377o	9	P(19)
5o864	5	P(18)	53888	9	P(2o)
51o8o	5	P(2o)?	54oo4	9	P(21)
51224	6	P(15)?	54o62	1o	P(15)?
51324	6	P(16)	54284	1o	P(17)?
51426	6	P(17)	54514	1o	P(19)
51536	6	P(18)	54926		?
51642	6	P(19)	55o4o		?
51752	6	P(2o)?	55272		?
51904	7	P(15)?	5539o	11	P(2o)
52oo6	7	P(16)	55932	12	P(18)?
52222	7	P(18)	56o52	12	P(19)