CLM - P 525

CLM - P 525

UKAEA

Preprint

OPTICALLY PUMPED LASER ACTION IN DIDEUTEROACETYLENE

H N RUTT
J M GREEN

CULHAM LABORATORY
Abingdon Oxfordshire

1978

CLM-P 525

OPTICALLY PUMPED LASER ACTION IN DIDEUTEROACETYLENE

H.N. Rutt and J.M. Green
UKAEA Culham Laboratory, Abingdon, Oxon OX14 3DB UK.

ABSTRACT

Optically pumped laser action has been observed in dideuteroacetylene. A total of fifteen output wavelengths in the 17.4 to 20.5 micron region are produced, pumped by seven CO_2 laser lines. The laser shows a number of unusual features, in particular pumping on hot bands and formally forbidden transitions and operation at high pressure (65 Torr). Output energies were ~ 2 mJ from an unoptimised system. The temperature and pressure dependence of the output energy has been measured for typical lines. The time dependence of the output pulse varies markedly between the various output lines and suggests that one transition is Raman in origin.

(To be published in Optics Communications)

February 1978

1. INTRODUCTION

Molecular lasers operating on vibrational transitions pumped by a CO_2 laser form a useful source of mid-infrared radiation, and also provide new information on the spectroscopy and kinetics of the molecule concerned. In contrast to the very numerous pure rotational far-infrared lasers, relatively few lasers of this type have been reported, some of the best known being OCS,(1) CF_4,(2) CF_3I,(3) NOCl,(2) NH_3(4,5) and C_2H_4.(6) We report here for the first time laser action in dideuteroacetylene, C_2D_2. This molecule displays a number of interesting features and provides 15 new wavelengths between 17.4 and 20.4 μm.

2. EXPERIMENTAL

The $^{12}C_2D_2$ used in these experiments was prepared by the action of heavy water on commercially obtained calcium carbide, which was de-gassed under vacuum prior to use. The C_2D_2 was dried over phosphorus pentoxide but not otherwise purified. The only impurities detected in the infrared absorption spectra were traces of C_2HD, D_2S and HDS and a very small amount of C_2H_2.

A modified Lumonix model 203 line tunable TEA CO_2 laser was used to pump the gas. The optically pumped laser system comprised a 12.5 mm square internally polished copper pipe one metre long, terminated at one end by a 10m radius internal mirror with a 1 mm diameter output coupling hole, and at the other end by a Brewster's angle potassium bromide window. This window was orientated so as to favour laser oscillation polarized parallel to the linearly polarized pump laser beam, which was introduced into the square pipe slightly off axis. The pipe could be cooled to 77 K.(7) The laser cavity was 2.3 m long, the other mirror being of 5m radius.

Output wavelengths were measured with a 1 metre monochromator with a resolution ~ 0.2 cm^{-1} around 500 cm^{-1}. The laser frequencies could be measured to ± 0.2 cm^{-1} for strong lines and to ± 1 cm^{-1} for weak lines. The accuracy was limited by the reproducibility of the laser power. Second order grating responses and scattered light were eliminated with 12 μm long wave pass filters and time resolved measurements made with a mercury cadmium telluride detector having a 50 ns response time.

C_2D_2 pressures were measured with a capacitance manometer.

3. ASSIGNMENTS

The bending mode levels of C_2D_2 relevant to laser action are shown in Fig.1. Extensive high resolution spectral data are available for many bands in C_2D_2 (8-11) and made possible the prediction of optically pumped laser action.

The $(\nu_4 + \nu_5)$ band overlaps the $^{12}C^{16}O_2$ 9 μm laser band, and the Σ_u^+ component of this band has been analysed in detail in (10). Transitions to the Σ_u^- and Δ_u levels are forbidden from the Σ_g ground state, but data for the Δ_u level is available from the hot band absorpiton $(\nu_4 + \nu_5)\Delta_u \leftarrow \nu_4\Pi_g$ given in (11). Two hot bands occur in the same spectral region, $(2\nu_5 + \nu_4)\Pi_g \leftarrow \nu_5\Pi_u$ and $(2\nu_4 + \nu_5)\Pi_u \leftarrow \nu_4\Pi_g$; and these bands have also been analysed in (10). Two further hot bands should exist as shown in Fig.1, but have not so far been observed. From the hot band pumped Π upper levels two possible laser transitions exist, to the $\Sigma_{u,g}^+$ and $\Delta_{u,g}$ levels.

Lines pumped from the ground state will generally increase in intensity at low temperature due to the reduction of the population in the lower laser level, although this effect may to some extent be reduced if the levels are of high J. Where the lower level of the pump transition is vibrationally excited, a rapid decrease in laser output with decreasing temperature is expected for high J levels, while for low J levels this effect may be compensated by rotational redistribution, reduced lower laser level population and reduced rotational relaxation. The laser transitions from hot band pumped levels evidently have higher vibrational quantum numbers than the ground state pumped lines. This leads to increased stimulated emission cross sections, and lower minimum pump level populations for threshold gain.

The assignment of the transitions pumped by R(12), P(24) and P(36) is straightforward, as shown in Table 1. The lines pumped by P(24) only lase at low temperature (< 230 K) owing to the high value of the rotational partition function (245) at room temperature.

The strong lines pumped by R(20) persist at 200 K but cannot be assigned to the $(\nu_4+\nu_5)\Sigma_u^+$ to $(\nu_4)\Pi_g$ band or to transitions pumped on a hot band. Although the ground state to $(\nu_4+\nu_5)\Delta_u$ transition has not previously been observed in absorption, calculations from the constants given in (11) show the assignment given in Table 1 to be consistent. Direct observation of this transition is reported in (12).

The lines pumped by R(14) are unassigned. No coincidence exists between R(14) and any published absorption line of C_2D_2. This line rapidly decreases in output power with decreasing temperature, which strongly suggests hot band pumping. The most likely origin for this band is pumping to one of the Π states of $(2\nu_4+\nu_5)$ or $(2\nu_5+\nu_4)$ for which no constants are available. The lines form a series for P(25), Q(24), R(23) approximately with $\Delta B \sim 5\times10^{-3}$ which is a reasonable value for this molecule.

There are two hot band transitions close to the CO_2 P(38) pump line, but the output wavelengths identify the transition pumped as P(4) of $(2\nu_5+\nu_4)\Pi_g \leftarrow \nu_5$. Laser transitions occur to two lower vibrational levels, Δ_u and Σ_u^+ of $\nu_5+\nu_4$.

Again two hot band transitions lie close to the CO_2 P(26) pump. One of these, R(2) of $(2\nu_5+\nu_4) \leftarrow \nu_5$ would give rise to the same upper level as the P(38) pump line and hence the same output frequencies. The pump transition is therefore identified as the Q branch of $(2\nu_4+\nu_5) \leftarrow \nu_4$, which will only have appreciable strength at low J as it is a $\Pi \rightarrow \Pi$ transition.$^{(13)}$ The output wavelengths correspond to transitions to the Σ_g^+ lower level, and are of low J as expected.

4. LASER CHARACTERISTICS

Optically pumped laser action is obtained very easily in C_2D_2, and the various lines are shown in Table 1. In comparison with the CF_4 optically pumped laser (ref.1) under conditions optimized for this gas, C_2D_2 gave output energies approximately three times larger on the strongest transition. On the strongest lines (those pumped by R(12)) the laser pulse energy is very reproducible even at room temperature, again in strong contrast to the CF_4 laser which, even under optimized conditions at low temperature, shows strong fluctuations in output.(7)

At room temperature thresholds were ~ 300 mJ for the R(12) pump line with 6 Torr of C_2D_2. Under the same conditions 2 J input produced ~ 2 mJ total output (P+R).

The temperature dependence of the C_2D_2 laser energy (all lines) was measured for the lines pumped by CO_2 P(24), R(12) and R(14), and the results are shown in Fig.2. For R(12) the slow rise with decreasing temperature is due to reduction in the lower level population partially offset by rotational redistribution in the ground state. The P(24) pumped transition on the other hand benefits from both these effects. The R(14) pumped transition almost certainly originates in a hot band, but the decrease in laser energy with temperature is partly due to the high J (~ 23) of the laser levels.

All the lines, with the exception of those pumped by P(24) could be obtained at room temperature. The transition pumped by P(26) was very weak and sporadic in operation, as were the two lines marked as weak in Table 1 pumped by P(38). All the other lines were readily obtained and oscillated reliably.

P(38) and P(26) have close coincidences with C_2D_2 hot band lines as shown in Table 1, and the lasing frequencies strongly support the hot band assignment. However, the laser output on these lines increased on cooling to 200 K; if the assignments are correct it must be assumed that this temperature behaviour reflects the low J quantum numbers of the levels involved. In view of this temperature dependence these hot band assignments require further confirmation.

The pressure dependence of the total output energy for the R(12) pumped lines is shown in Fig.3. C_2D_2 lases over an exceptionally wide pressure range of up to 65 Torr, as compared to 12 Torr for OCS and to 23 Torr for CF_4 measured in the same apparatus.(7) Diode laser measurements on C_2D_2(12) show the self pressure broadening coefficient to be ~ 12 MHz/Torr, so that at 65 Torr strong rotational relaxation is occurring.

5. OUTPUT PULSE SHAPES

Typical pulse shapes are shown in Fig.4.

The lines pumped by R(12) lase within 65 ns of the pump pulse, which is ~ 5 cavity round trips. This indicates very high gain, and we conservatively estimate a gain coefficient of 2.5% cm^{-1}. Laser action was maintained throughout the weak 'tail' of the CO_2 pump pulse for up to 2 μs.

All the other lines except those pumped by R(20) show much longer delays indicative of lower gain. For the various lines delays range from 200 ns - 1.5 μs, with pulse widths of 500 ns - 1.0 μs. The two lines pumped by R(20) show markedly different behaviour. Laser action is nearly time coincident with the CO_2 laser gain switched spike, and terminates at the end of the spike. This is probably indicative of Raman laser action.

6. CONCLUSIONS

The C_2D_2 laser shows a number of features which have not previously been observed: laser action on lines of very low J (Q(1)), laser action on transitions pumped by a forbidden transition ($\Delta \ell = 2$) and laser action pumped on a vibrational hot band.

Aside from these interesting spectroscopic features, substantial output energies have been obtained (2 mJ) in an unoptimized system. With increased output coupling we anticipate large output powers being obtainable.

The long duration pulses produced by R(12) pumping combined with high pressure operation strongly suggest that inversion of the entire $(\nu_4 + \nu_5)\Sigma_u^+$ manifold will be possible. This would permit grating runing and operation on some 40 lines in the 17-20 μm region. A second set of lines would be available if the $(\nu_4 + \nu_5)\Delta_u$ state can be similarly inverted or collisionally pumped from the Σ_u state. Collisional pumping of $2\nu_4$ and $2\nu_5$ from $\nu_4 + \nu_5$ may also be possible.

Other close coincidences exist between various $^{12}C_2D_2$ absorption lines and $^{12}C^{16}O_2$ laser lines, and it is probable that other transitions can be obtained (eg with a longer cell and higher pump energies). Further coincidences exist between $^{12}C^{16}O_2$, $^{13}C^{16}O_2$ and $^{12}C^{18}O_2$ laser lines with $^{12}C^{13}CD_2$ and $^{13}C_2D_2$, and would provide a considerable number of laser lines in the 17-21 μm region.

ACKNOWLEDGEMENTS

We would like to acknowledge the help of Dr. D.N. Travis in assigning some of the laser transitions, and for many useful discussions.

REFERENCES

1. SCHLOSSBERG. H.R., LETTERMAN, H.R. - Appl.Phys.Lett.26, 316 (1975)
2. TIEE. J.J., WITTIG. C. - Appl.Phys.Lett.30, 420 (1977)
3. PROSNITZ. D., JACOBS, R.R., BISCHEL, W.K., RHODES, C.K. - Appl.Phys. Lett.34, 221 (1978)
4. JACOBS. R.R., PROSNITZ, D., BISCHEL, W.K., RHODES, C.K. - Appl.Phys. Lett.29, 710 (1976)
5. CHANG, T.Y., McGEE, J.D. - Appl.Phys.Lett.28, 526 (1976)
6. CHANG, T.Y., McGEE, J.D. - Appl.Phys.Lett.29, 725 (1976)
7. GREEN, J.M. - European Conference on Optical Systems and Applications, Brighton, April 1978.
8. POLLARD, K.P., GHERSETTI, S., RAO, K.N. - J.Mol.Spec. Vol.30, pp.146 (1969)
9. HURLOCK. S.C., GHERSETTI, S., RAO, K.N. - Mem.Soc.Roy.Sci.Liege Collect. Vol.1, No.2, pp.67 (1971)
10. GHERSETTI. J. PLIVA, RAO, K.N. - J.Mol.Spec.Vol.38, pp.53 (1971)
11. BALDACCI, A., GHERSETTI, S., HURLOCK, S.C. - J.Mol.Spec.Vol.42, pp.327 (1972)
12. RUTT. H.N., TRAVIS, D.N. - to be published in J.Phys.B 11 (1978)
13. HERZBERG. G. - Molecular Spectra and Molecular Structure Vol.II, pp.390 Van Nostrand 1945.

TABLE 1

OBSERVED LASER TRANSITIONS IN C_2D_2

$^{12}C^{16}O_2$ Pump	$^{12}C_2D_2$ Absorption		Laser Line	Lower Level	Observed ν cm^{-1}	Strength
R(12)	R(17)	$(\nu_4 + \nu_5)\Sigma_u^+ \leftarrow GS$	P(19)	ν_4	499.7	VVS
			R(17)	ν_4	562.6	VS
R(20)	R(16)	$(\nu_4 + \nu_5)\Delta_u \leftarrow GS$	P(18)	ν_4	508.4	M
			R(16)	ν_4	567.8	M
P(24)	R(0)	$(\nu_4 + \nu_5)\Sigma_u^+ \leftarrow GS$	P(2)	ν_4	527.4	S
			Q(1)	ν_4	530.6	S
P(36)	P(6)	$(\nu_4 + \nu_5)\Sigma_u^+ \leftarrow GS$	P(6)	ν_4	520.7	S
R(14)	R(23)?	Hot Band	P(25)?	?	489.2	W
			Q(24)?	?	535.7	W
			R(23)?	?	573.1	W
P(38)	P(4)	$(2\nu_5 + \nu_4)\pi_g \leftarrow \nu_5$	P(4)	$(\nu_4 + \nu_4)\Delta_u$	518.9	VW
			P(4)	$(\nu_5 + \nu_4)\Sigma_u$	525.6	VVW
			Q	$(\nu_5 + \nu_4)\Delta_u$		
			Q	$(\nu_5 + \nu_4)\Sigma_u$	532.2	W
P(26)	Q	$(2\nu_4 + \nu_5)\pi_u \leftarrow \nu_4$	Q (?)	$(2\nu_4)\Sigma_g$ (?)	527.1	VVW
			P(3)	$(2\nu_4)\Sigma_g$	522.7	VVW

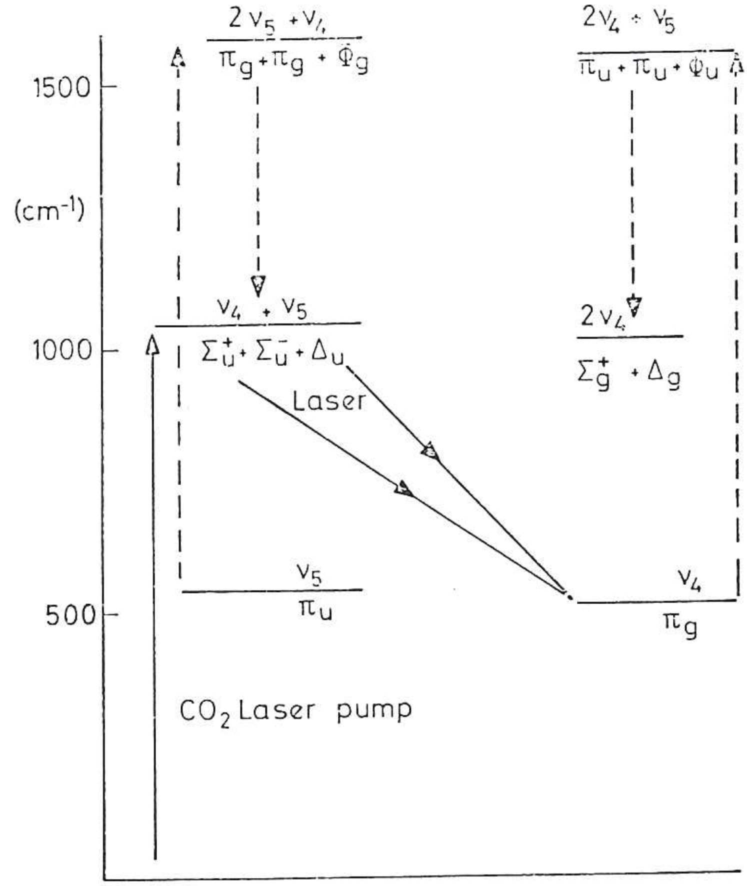

Fig. 1 Energy levels in C_2D_2

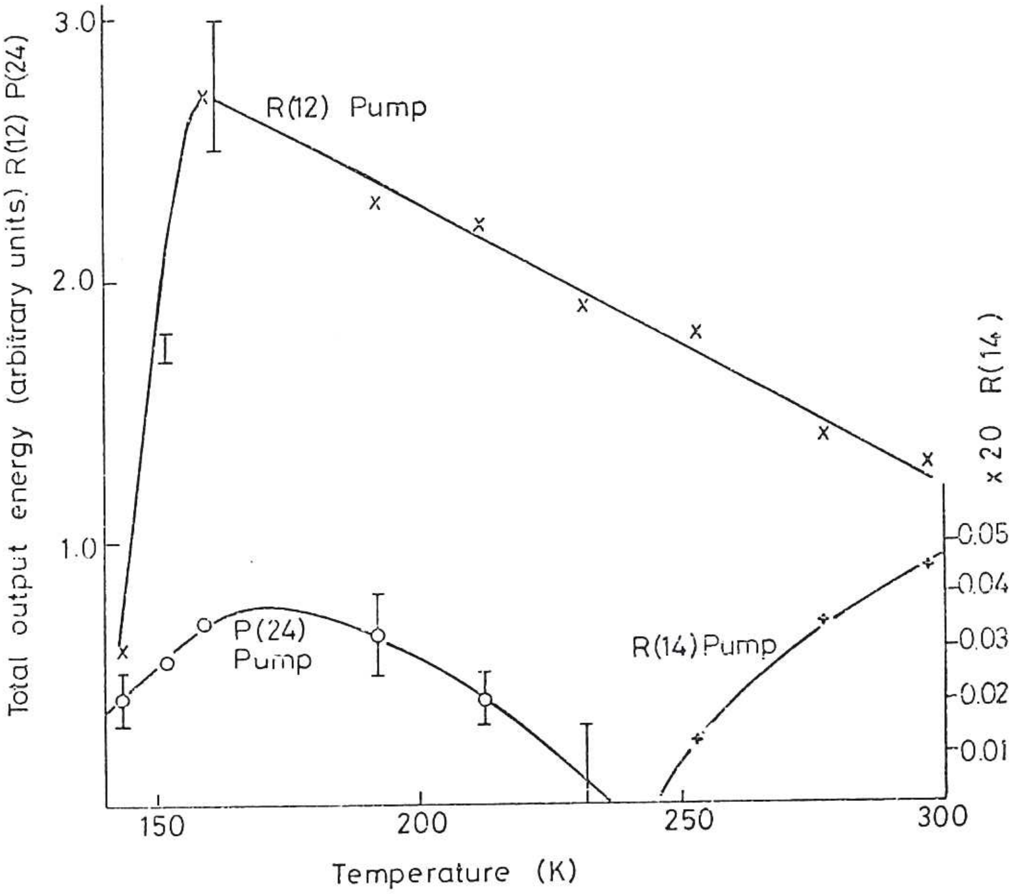

Fig. 2 Temperature dependence of the output pulse energy. 4.5 Torr C_2D_2

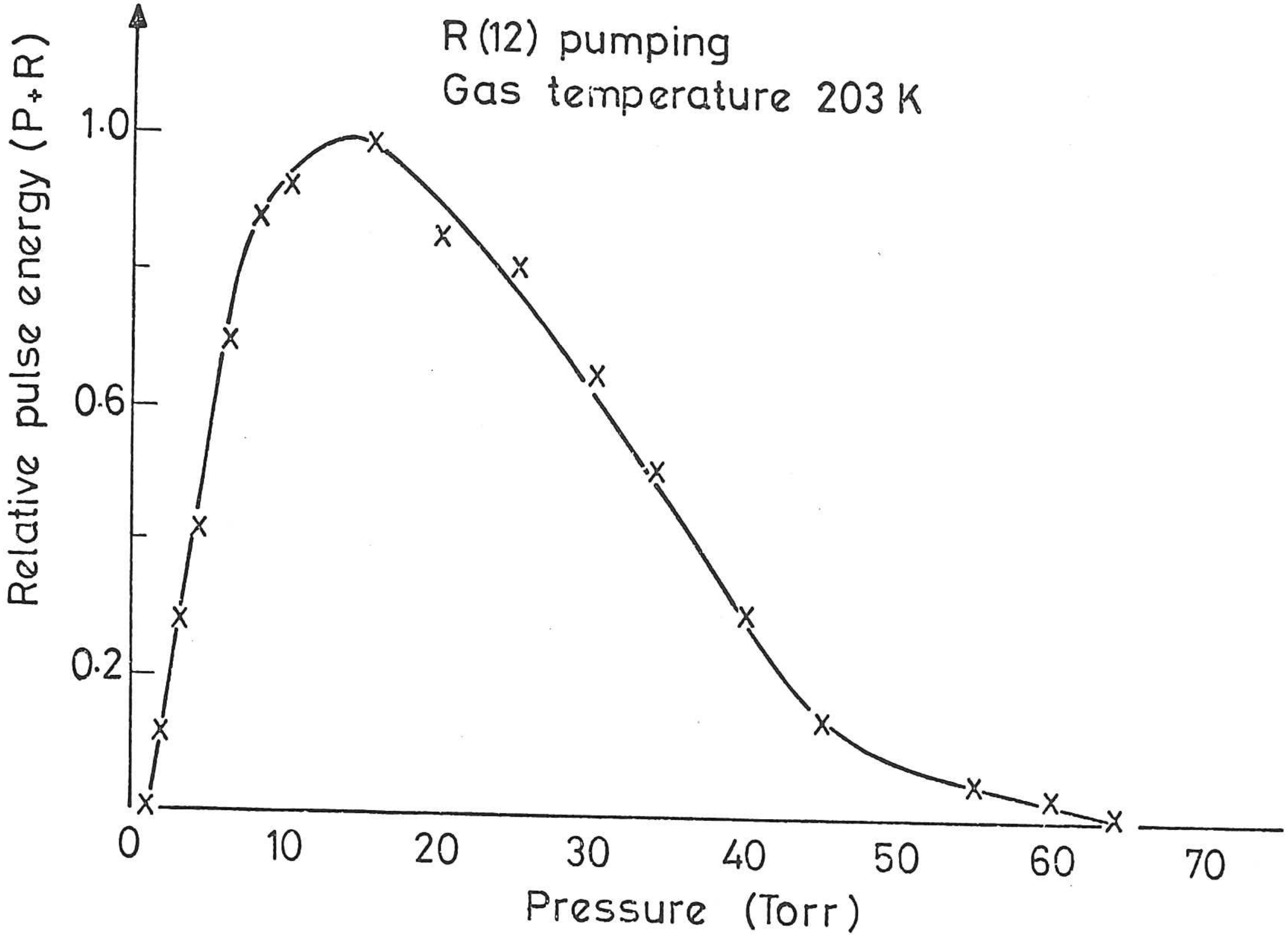

Fig. 3 Pressure dependence of output pulse energy

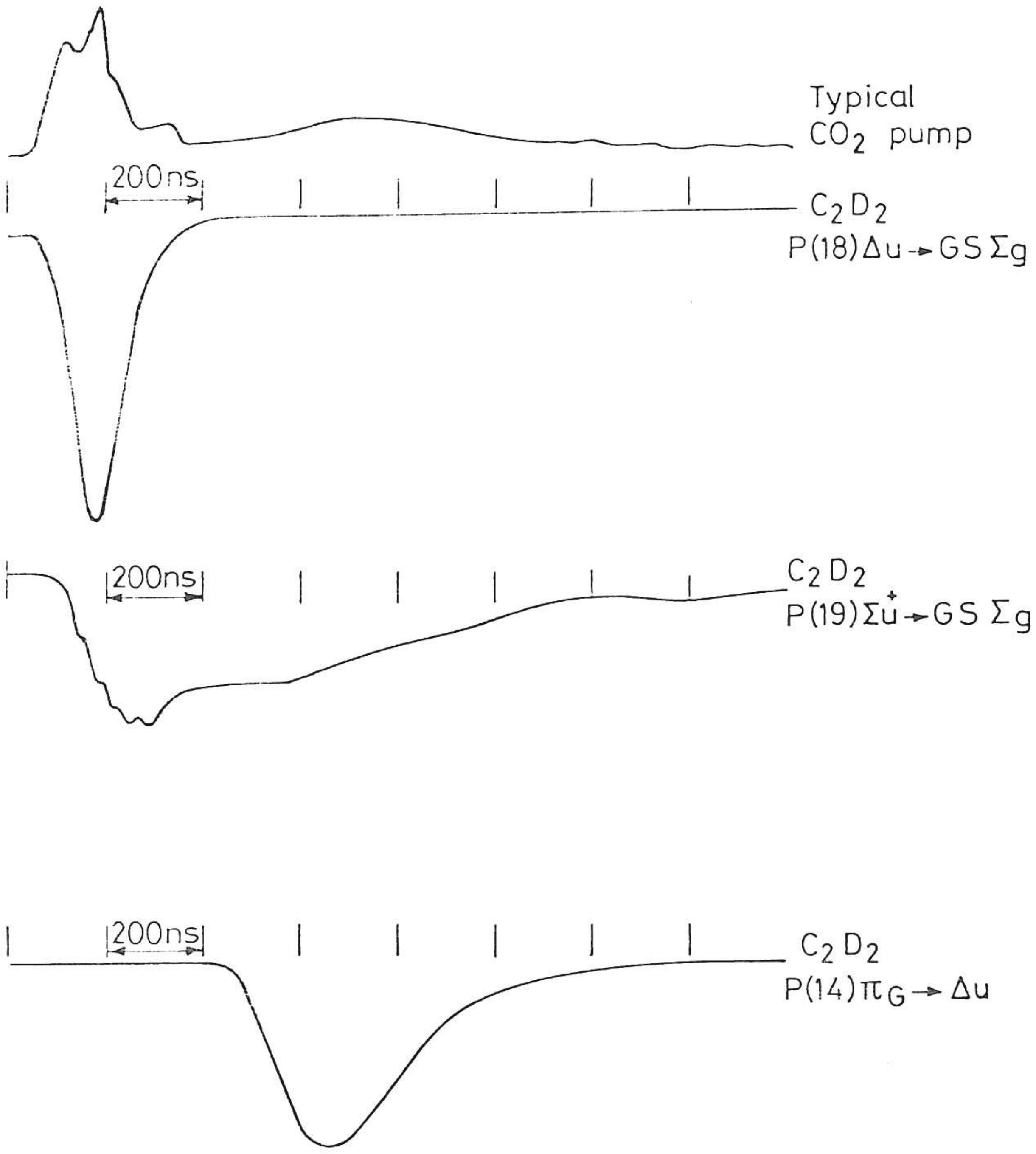

Fig. 4 Time dependence of the C_2D_2 laser output