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TITLE: STIMULATED RAMAN SCATTERING AND FOUR-WAVE MIXING IN CO_2-PUMPED PARA-H_2

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STIMULATED RAMAN SCATTERING AND FOUR-WAVE MIXING

IN CO_2-PUMPED PARA-H_2

J. L. Carlsten and N. A. Kurnit

(Text of talk to be presented by J. L. Carlsten at Topical Meeting on Infrared Lasers, Los Angeles, CA, Dec. 3-5, 1980)

We have for some time been following an alternate approach to the Raman shifting of CO_2 radiation than the one to be described today, namely the amplification in a hollow dielectric waveguide of Stokes radiation provided by either a tunable diode laser or an HF pumped OPO. The setup for these experiments, which was described at the CLEOS meeting,[1] is shown in Fig. 1, which is relevant to the work to be discussed here. Basically, the Stokes source and CO_2 are combined with orthogonal polarization on a Ge beamsplitter and rendered oppositely circularly polarized by a KBr Fresnel rhomb, and then co-propagated through the amplifying medium, in this case contained in a 3m LN_2-cooled alumina waveguide. With this system, gain as high as e^9 on the peak of mode-locked spikes was observed, and with an HF OPO as input source, the system could be driven into pump depletion, but only for well-mode-locked pulses.

With the success of Trutna and Byer[2] and the Exxon group,[3] in using multipass refocusing cells, we began work on these also, and I will describe today our work both with a room temperature multipass cell (MPC) similar to the one studied by Trutna and Byer[2] and also with a LN_2-cooled MPC as has been studied extensively by the group at Exxon.[3]

In particular, the results listed in Fig. 2 obtained with the multipass cell whose characteristics are given in Fig. 3 will be discussed. In addition to the gain measurements with the diode, we have obtained fully-depleted pump pulses with an OPO input, and have observed two-frequency depletion using two-pump pulses and one Stokes input, which is initiated by four-wave mixing. Such two-frequency operation has been reported by Rabinowitz.[4] We have also obtained pump depletion starting from noise with a strong 9-μm pump.

In a recent experiment we have used a microwave-shifted[5] output of a CF_4 laser[6] as an injection seed source into the room temperature MPC which allowed us to make gain measurements as a function of probe frequency. We have also run our MPC at 78 K which with the increase in gain allowed us to obtain full pump depletion with no injected seed source.

In all of these experiments the process involved is stimulated rotational Raman scattering in para-H_2. Figure 4 shows an energy level diagram of the relevant rotational levels in the lowest vibrational state of para-H_2. Scattering occurs from the J = 0 to the J = 2 rotational state which gives a Raman shift of 354.36 cm^{-1}. By using para-H_2, from the blowoff of LH_2, one eliminates the odd rotational levels and thereby increase the gain for the J = 0 to J = 2 Raman transition.

Figure 5 shows the setup we used for measuring the gain using a 17-μm diode. The previously mentioned waveguide was used as a pre-amplifier so that we could monitor the input signal to the multipass cell and the amplified output on similar, moderately fast (~100 ns) HgCdTe detectors. Also, in this way, the Stokes input spectrum to the multipass cell has been tailored to that of the free-running CO_2, with the exception that the bandwidth of the room temperature H_2 will not support pulses as narrow as those which the low temperature, higher pressure waveguide can produce.

Pictures taken with a high speed Cu:Ge detector of the pulses amplified by the room temperature cell alone (Fig. 6) indicate structure which is somewhat more highly peaked than that of the CO_2, but not greatly so, indicating that the gain cannot follow the very fast CO_2 spikes, as it can at lower temperature and higher pressure. We have therefore used an averaged value for estimates of the peak power, although this may somewhat underestimate the effective power. We have also seen comparable outputs with a smoothed CO_2 input pulse, but with less reliability, which we believe is due to the greater accuracy needed in diode tuning th the narrow-line CO_2 source.

Figure 7 gives a comparison of the gain measured with the diode as well as that obtained from the buildup from noise with the 9-μm pulse, with the theoretical gain, calculated from what are believed to be reliable values of the anisotropic part of the polarizability[7] and Raman linewidth,[8] which has been measured to be 23% greater in room temperature para-H_2 than the value normally quoted for normal-H_2. The gain at low temperature is not as accurate as that at room temperature because the low-temperature linewidth has not been measured. Calculations based on the work of Van Kranendonk[9] indicate that the linewidth at 77 K differs from that at room temperature only by the difference in density, but this theory has been shown[8] to give the wrong dependence of linewidth on ortho-para concentrations unless an arbitrary choice is made for the collision time. It is doubtful that this or any other theory we are aware of[10] can be trusted to give a good a-priori value of the low temperature linewidth; however the assumption that $\Gamma \sim N$ is probably valid to withi . 20 or 30%.

The experimental number has been extracted using the mode-overlap result used by Rabinowitz,[3] and assumes the pump and Stokes are both given by lowest order modes. These assumptions do not accurately account for diffraction losses due to the non-uniform gain profile, and hence tend to overestimate the gain;[11] also, the gain is lower for any higher-order mode contained in the pump.

Figure 8 shows the setup used with the HF OPO. In this case, the Stokes frequency was fairly strongly absorbed by the CO_2 in the amplifier, and it was necessary to combine the beams after the amplifier. Figure 9a shows a 10 μm CO_2 pump pulse observed on a photon drag detector after the multipass cell with and without the OPO, together with a typical Stokes output pulse. One can see that the pulse is in fact taken down to the baseline over a large portion of the pump pulse. Energy outputs of up to 800 mJ (accounting for optics losses) have been measured in this way.

Figure 9b shows the effect of adding a second CO_2 laser frequency, which happens to be a 9-μm line derived from the same CO_2 oscillator.[12] The top trace shows the 9-μm pump, the middle trace the 10-μm pump, which is even more strongly depleted than in the corresponding picture without the 9-μm input, and finally, the 9-μm depleted pump, which is also strongly depleted even though it is not seeded (and at this pump power would not generate any Stokes output without the 10-μm pump and its Stokes). This behavior is expected on the basis of the theory of four coupled waves interacting with the medium,[13] as we can see from Fig. 10.

Under the assumption of no pump depletion and no dispersion (which is strictly valid in this case since the coherence length is kilometers for any combination of CO_2 lines), the equations for four-wave-mixing (with phase set to that which maximizes the growth of the weakest wave) yield solutions in which the Stokes waves grow together with an exponential gain factor which is the sum of the gain from the two waves. They differ only in the pre-exponential factor which is determined by which wave is seeded. In Fig. 11, these solutions are written in terms of the output intensities in the high gain limit, and we see that the outputs are in the ratio of the input intensities, aside from frequency factors. This behavior is expected to be carried on into saturation, and this has been verified through analytic solutions obtained by Jay Ackerhalt,[14] as indicated in Fig 11 a.

Figure 12 presents some data of output energy near 14 μm and 16 μm as a function of the 9-μm pump energy for three values of 10-μm pump with only the 10 μm seeded. For strong 10-μm pump, the corresponding output at ~16 μm is relatively constant over the whole range (the dip at high 9-μm energy is due to the 10 μm output of the Lumonics amplifier becoming weaker for strong 9-μm pulses.) Even at low 9-μm pump intensity a large fraction of 9 μm is converted to 14 μm. As the 9-μm intensity is increased, this fraction also increases. For smaller values of 10-μm energy, little 14 μm is produced until the 9-μm energy is made large, and the 16-μm energy also shows a corresponding increase; that is, the 9 μm becomes the pulse which drives the system, even though the 10 μm is the one which is seeded.

Figure 13 shows a 9 μm depleted pulse with no input Stokes source. This laser happened to produce a relatively short pulse on this line, with essentially no energy in a tail. Thus the peak power was considerably higher for the 9-μm line than for the 10, and the frequency factor in the gain is also in its favor. The threshold for full depletion near the peak of the pulse was also near the threshold for gas breakdown in the cell, although this may have been caused by dust in the cell. Our breakdown threshold continuously got higher as a function of time after having the cell open.

Figures 14-16 are photos of our MPC which can be operated at either 78 K or 300 K. Running this cell at room temperature, we measured the amplification of a microwave shifted CF_4 laser as a function of microwave shift. The experimental apparatus is diagramed in Fig. 17. Basically the CF_4 replaced the OPO of previous experiments. However in this case the CF_4 laser was single mode and the microwave shifts allowed fine frequency tuning of the probe beam. The results of this experimental measurement of the gain vs frequency of the probe is shown in Fig. 18. With existing microwave equipment, we were able to tune in the region of 0.95 to 1.55 GHz from Raman resonance with the center of the CO_2 mode distribution and more recently, in a region believed to be close to line center. As can be seen from Fig. 18, the gain increased with some modulation as we tuned nearer to resonance. The modulation is due to the longitudinal mode structure of the CO_2 pump, which was run at 1 J to keep below saturation. The gain at line center for this experimental setup using single mode lasers is expected to be 10^{10}. A narrow peak, with relatively low gain, was seen with a nominally single-longitudinal-mode laser using an intracavity gain cell. It is quite possible that this was due to a weakly oscillating mode and that we are not yet tuned into exact Raman resonance.

Ackerhalt[14] has analyzed the amplification of an off resonant probe with a multimode pump laser and finds that for a probe that is tuned into Raman resonance with the nth mode of pump, the total Stokes output is

$$I_s = I_s^o \frac{I_{p,n}}{I_p} \exp(\alpha I_p z)$$

as shown in Fig. 19, where $I_{p,n}$ is the intensity of the nth mode of the pump and I_p is the intensity summed over all the longitudinal modes of the pump. Thus we see that the profile given in Fig. 19 is basically a measure of the CO_2 pump longitudinal mode distribution, and that even at line center the amplification will be less for the broadband pump due to this prefactor than for a single mode pump.

Figure 20 shows the experimental setup used for the cold cell operation. In this case the cell was cooled with LN_2. Figure 21 shows a typical oscilloscope tracing of the pump pulse before the MPC, the pump pulse after the MPC, and generated Stokes output for the MPC operating at 78 K with 39 passes. As can be seen, roughly half way up the CO_2 pump pulse, the stimulated Raman scattering is driven to saturation. In this case the pressure in the MPC was 150 torr para-H_2 and the CO_2 pump laser was run multi-longitudinal mode. In fig. 21a, similar depletion is shown with the cell operated at room temperature and 2J of input energy. We have also seen comparable depletion (Fig. 22 and 22a) with the CO_2 pump run single mode by the insertion of a low-pressure gain cell in the oscillator. Figures 23-24 show further comparison of the saturation with the single mode and multi-mode pump.

Figure 25 shows the effect of CO_2 pump energy on the CO_2 pump depletion. In this case the CO_2 laser was run single mode. The initial CO_2 pulse is shown by the dashed line. Pump depletion starts when the CO_2 pump is at the 1.28 J region. From this point on small increases in pump energy lead to large changes in depletion. We believe this is due to the inter-cell crossings discussed by Perry et al.[(4)]

Using the mode-overlap expression for gain, we have extracted an estimate of the plane wave gain coefficient, $\alpha = 8.6 \times 10^{-5}$ cm/MW, which is 12% lower than our theoretical value. We have estimated the transient effect using the calculations of Carman et al.[15]

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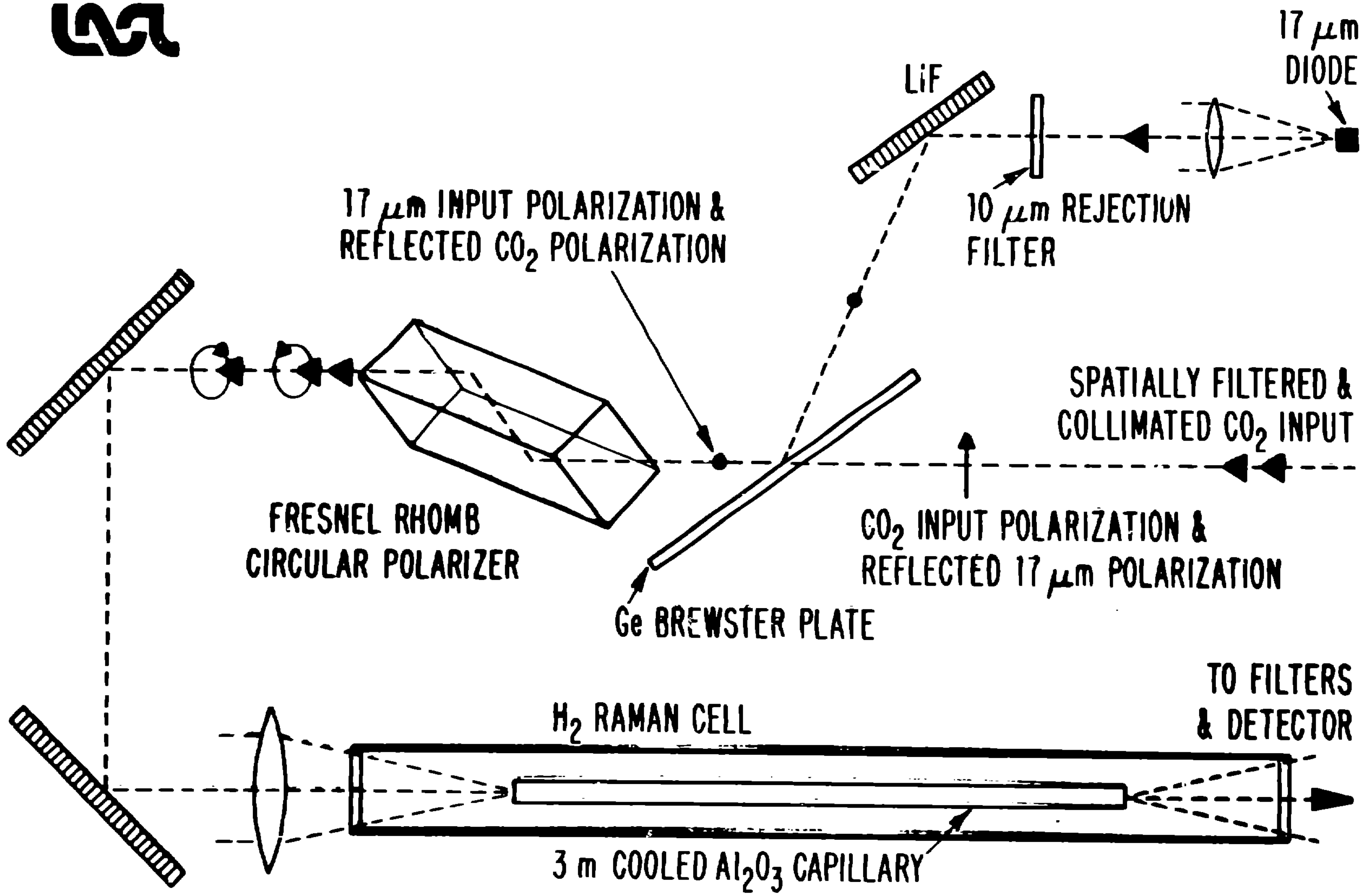
CO_2 RAMAN SHIFTING



AP-1-VG-2429B

H_2–RAMAN 300° K MULTIPASS CELL

RESULTS

- GAIN OF e^{16} OBSERVED WITH PRE-AMPLIFIED DIODE INPUT AND 10 MW CO_2 PUMP
- STRONG PUMP DEPLETION REACHED WITH $<$ 100 μJ OPO INPUT AT 10 MW CO_2 PUMP
  (WITH G.P. ARNOLD AND R.W. WENZEL)
- TWO-FREQUENCY SATURATED OUTPUT OBTAINED WITH 9 μm AND 10 μm INPUTS AND SINGLE STOKES INPUT VIA 4-WAVE MIXING
- PUMP DEPLETION STARTING FROM NOISE OBTAINED WITH 30 MW 9 μm PUMP
- GAIN MEASURED vs STOKES FREQUENCY (MICROWAVE SHIFTED CF_4 LASER)
  (WITH R. ECKHARDT, D.H. GILL, L. HAYNES AND J. TELLE)

H_2–RAMAN 78° K MULTIPASS CELL

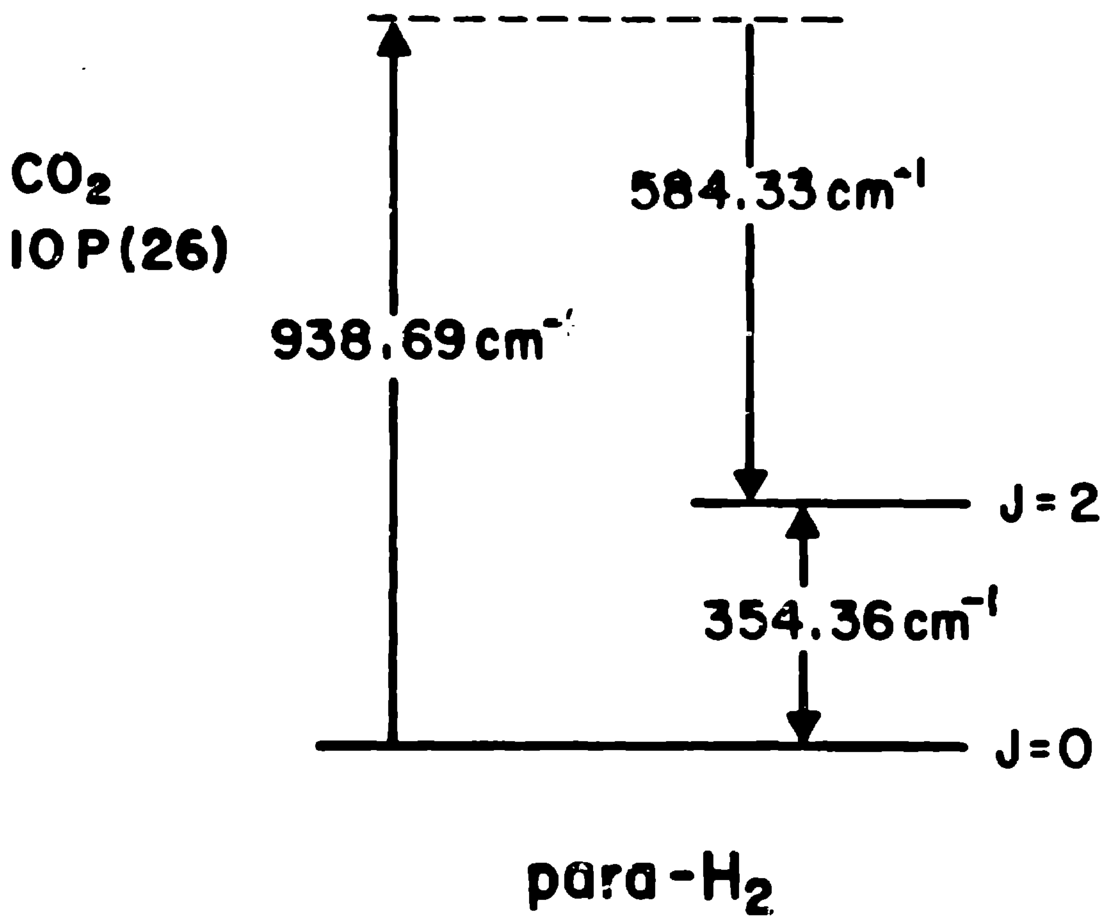
RESULT

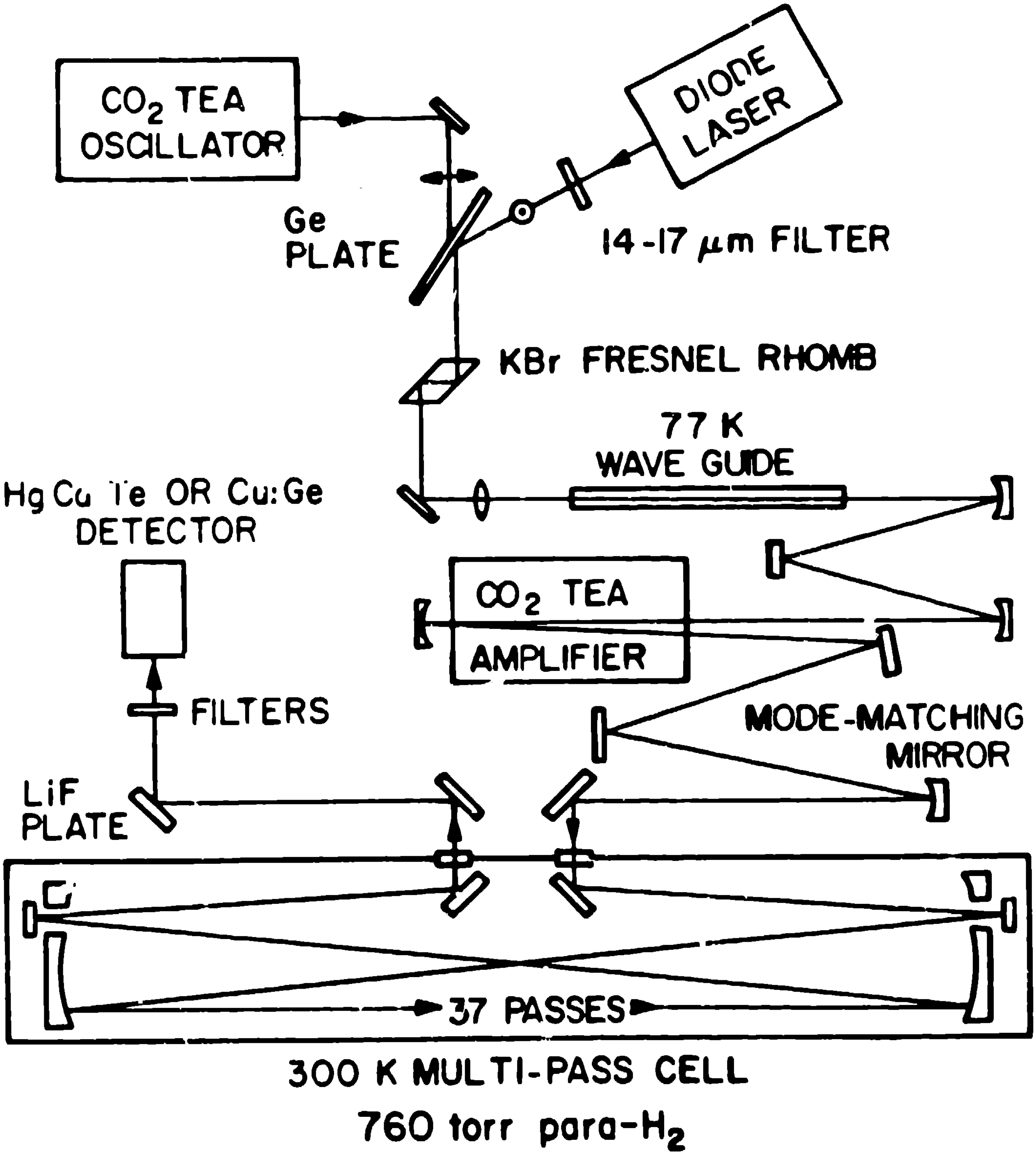
- STRONG PUMP DEPLETION STARTING FROM NOISE WITH 25 MW 10 μm PUMP

H_2 RAMAN MULTIPASS CELL CHARACTERISTICS

- OFF-AXIS SPHFRICAL INTERFEROMETER
- 37/39 PASSES, 98.7% REFLECTIVITY (50% THROUGHPUT)
- $R = 2$ m, $l = 3.47$ m, $\pi w_0^2/\lambda = 0.69$ m
- 1 atm. (TYPICAL) p-H_2 AT 300° K
- 200 torr (TYPICAL) p-H_2 AT 78° K

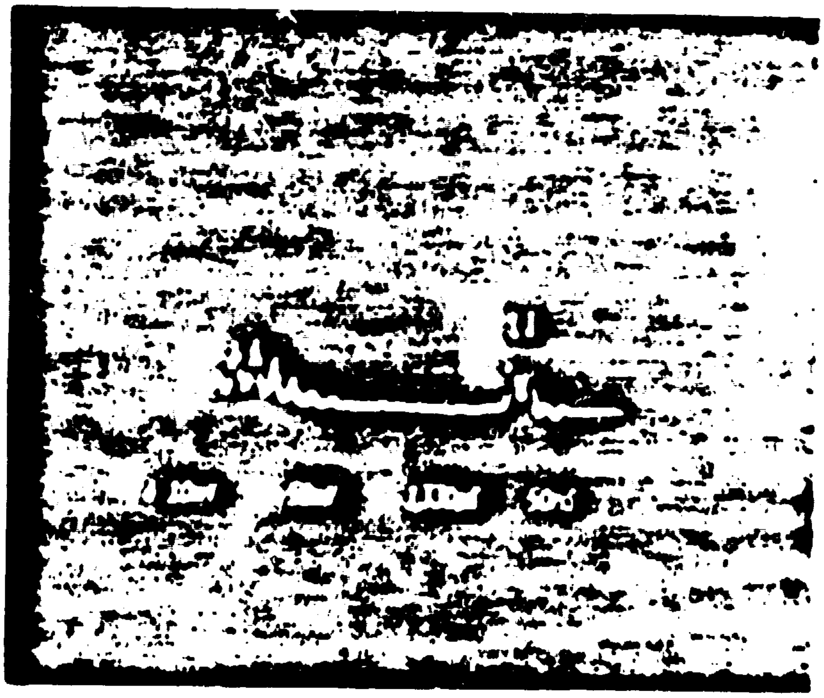
ENERGY LEVELS FOR PARA-H_2



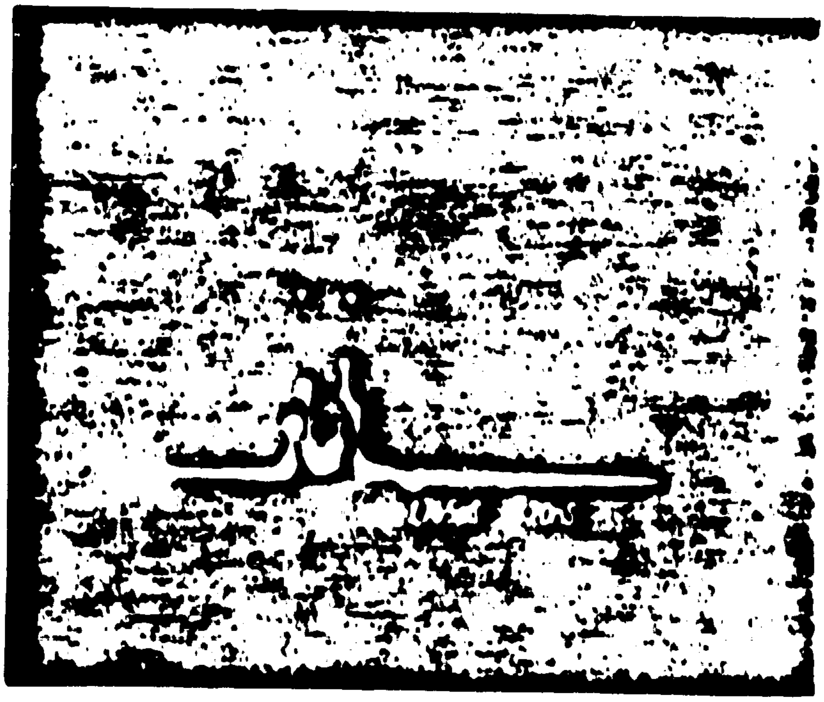

CO2 TEA
OSCILLATOR
DIODE
LASER
Ge
PLATE
14-17 μm FILTER
KBr FRESNEL RHOMB
77 K
WAVE GUIDE
Hg Cu Te OR Cu:Ge
DETECTOR
CO2 TEA
AMPLIFIER
FILTERS
MODE-MATCHING
MIRROR
LiF
PLATE
37 PASSES
300 K MULTI-PASS CELL
760 torr para-H2

AP-I-VG-6632

PUMP & STOKES



STOKES



H_2 RAMAN GAIN

$$g = \frac{64\pi^3 \chi_R}{\lambda_s c} \quad I_p = \alpha I_p \quad \text{(PLANE WAVE)}$$

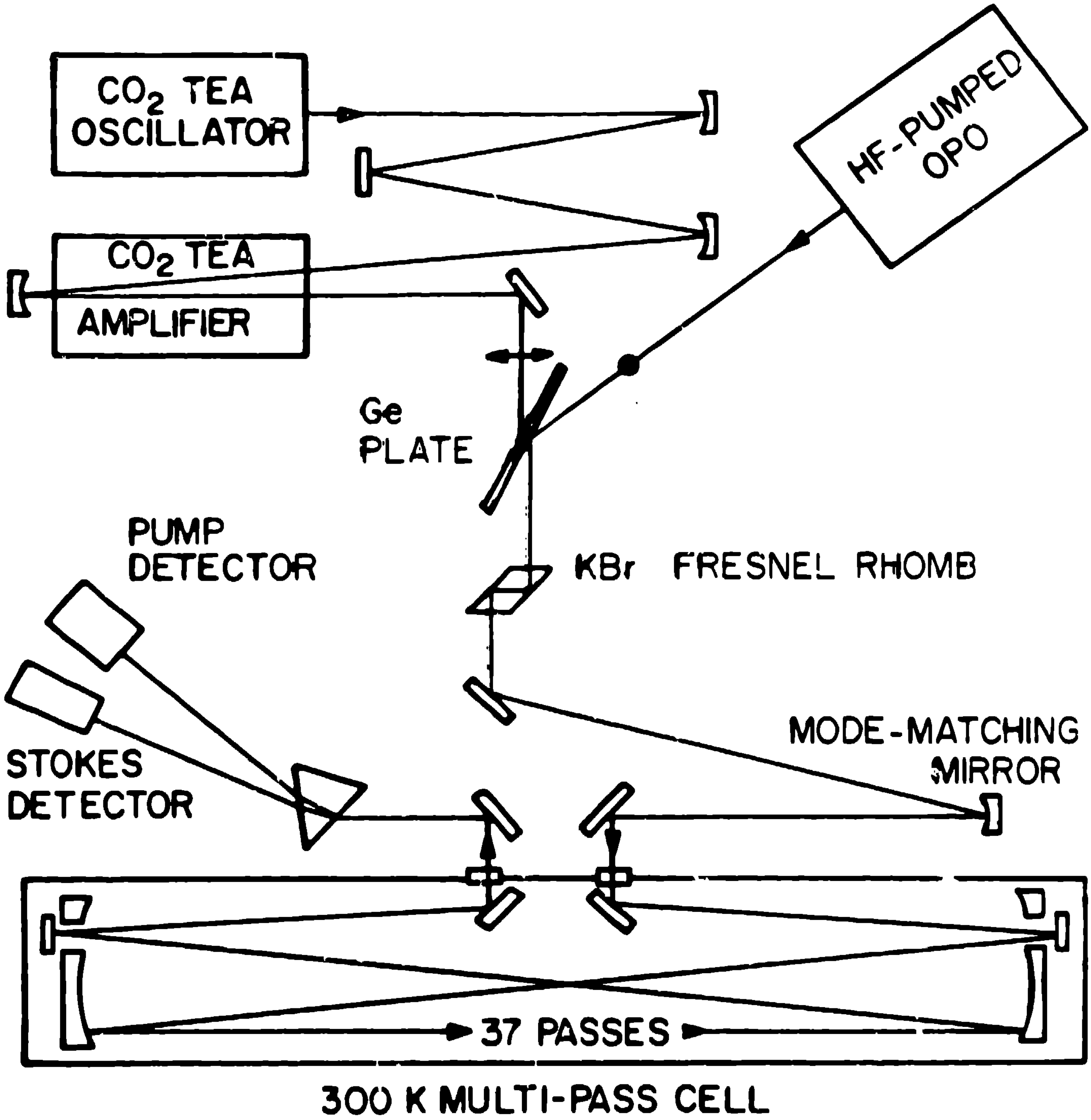
$$\chi_R = \frac{1}{5} \frac{(J+1)(J+2)}{(2J+1)(2J+3)} \gamma_{00}^2 \frac{N}{4h\Gamma}$$

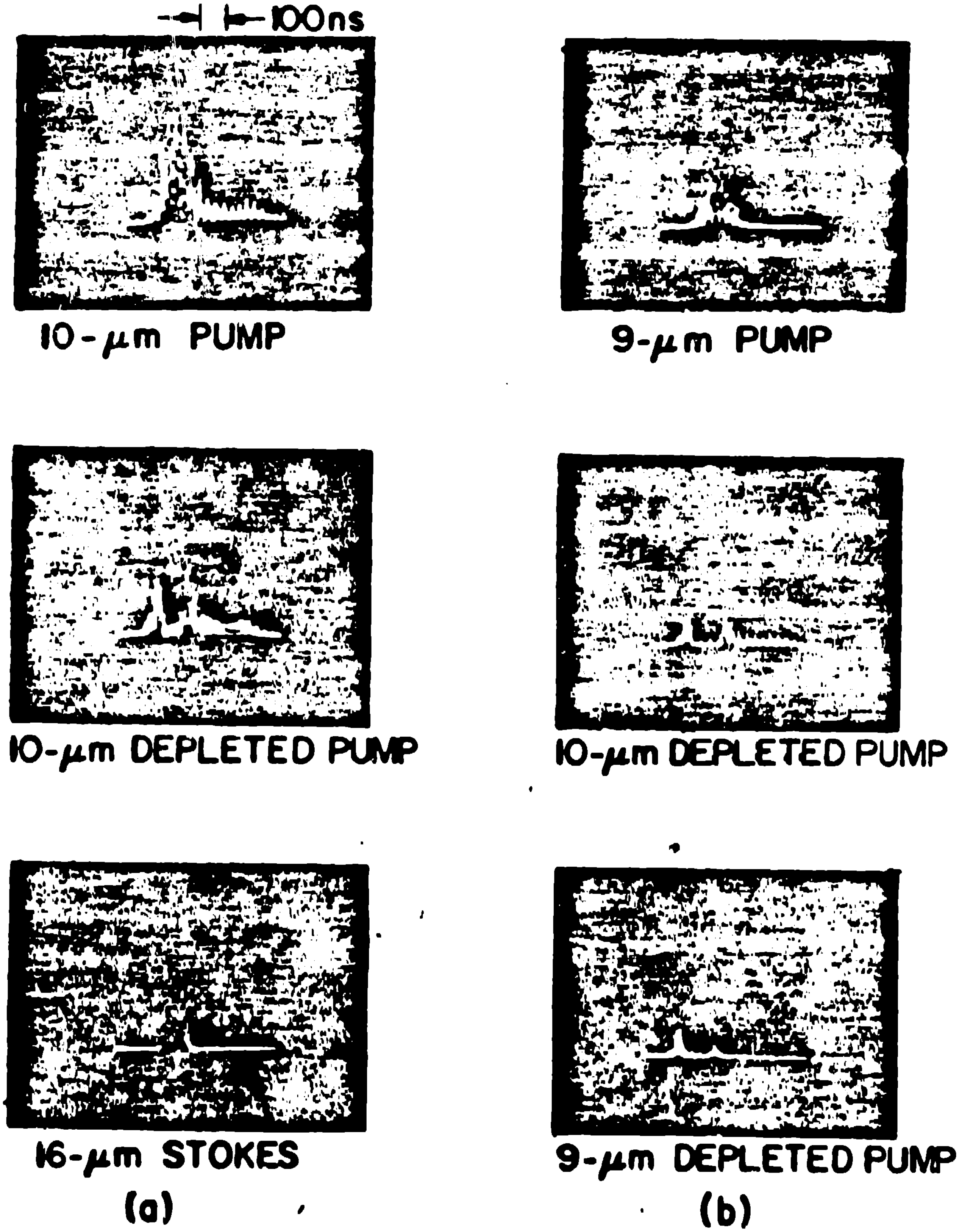
$$\int g\,dz = \frac{4\alpha P}{\lambda_s + \lambda_p} \left[\tan^{-1}(\xi\, b)\right] \frac{1 - r}{1 + r} \quad \text{(MULTIPLE FOCUS[1,9])}$$

	χ_R [a] (esu)	α_{theor} (cm/MW)	α_{exp} (cm/MW)
p-H_2 300 K $S_0(0)$	1.0×10^{13}	3.9×10^{5} ($\lambda_s = 17\ \mu m$)	3.0×10^{5} (DIODE, MPC)
		$\cancel{4.8 \times 10^{5}}$ ($\lambda_s = 14\ \mu m$)	$\cancel{2.2} \times 10^{5}$ 3.7 (GROWTH FROM NOISE, MPC)
77K	2.4×10^{13}	9.8×10^{5} ($\lambda_s = 16\ \mu m$)	7×10^{5} (OPO, WAVEGUIDE)
		9.2×10^{5} ($\lambda_s = 17\ \mu m$)	$\cancel{14} \times 10^{5}$ 8.4 (GROWTH FROM NOISE, MPC)

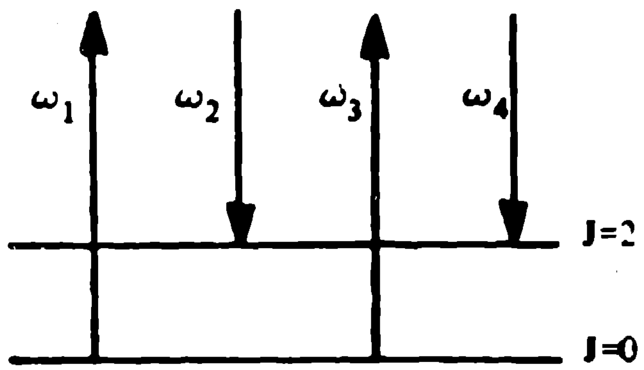
(a) WITH $\gamma_{00} = 0.30 \times 10^{24}\ cm^3$ (A. L. FORD AND J. C. BROWNE, Phys. Rev. A 7, 418 (1973))

$\frac{1}{2\pi} \Gamma\, [S_0(0), p\text{-}H_2] = 51$ MHz/AMAGAT (R. A. J. KEIJSER, J. R. LOMBARDI, K. D. VAN den HOUT, B. D. SANCTUARY, AND H. F. P. KNAAP, PHYSICA 76, 585 (1974))


CO2 TEA
OSCILLATOR
CO2 TEA
AMPLIFIER
HF-PUMPED
OPO
Ge
PLATE
PUMP
DETECTOR
KBr FRESNEL RHOMB
STOKES
DETECTOR
MODE-MATCHING
MIRROR
37 PASSES
300 K MULTI-PASS CELL


100ns
10-μm PUMP
9-μm PUMP
10-μm DEPLETED PUMP
10-μm DEPLETED PUMP
16-μm STOKES
(a)
9-μm DEPLETED PUMP
(b)

FOUR-WAVE MIXING



$$E(\omega_i) = E_i\, e^{i(\omega_i t - k_i z)}\, e^{-i\varphi_i}$$

$$\frac{dE_1}{dz} = -\frac{2\pi\omega_1}{\eta_1 c}\left[\chi''_R E_2^2 E_1 + \chi''_3 E_2 E_3 E_4\right]$$ (NO DISPERSION, PHASED FOR MAXIMUM GROWTH)

$$\frac{dE_2}{dz} = \frac{2\pi\omega_2}{\eta_2 c}\left[\chi''_R E_1^2 E_2 + \chi''_3 E_1 E_3 E_4\right]$$

$$\frac{d}{dz}(E_3, E_4) \rightarrow \text{SAME WITH } E_1 \leftrightarrow E_3,\ E_2 \leftrightarrow E_4$$

FOR E_1, E_3 = CONST. (NO PUMP DEPLETION), AND E_2 SEEDED,

$$E_2 = \frac{E_{20}}{1+g_1/g_3}\left(\frac{g_1}{g_3} e^{(g_1+g_3)z} + 1\right)$$

$$E_4 = \frac{E_1}{E_3}\,\frac{E_{20}}{1+g_1/g_3}\left(e^{(g_1+g_3)z} - 1\right)$$

WHERE

$$g_1 = \frac{4\pi\omega_2}{\eta_2 c}\,\chi''_R E_1^2, \quad g_3 = \frac{4\pi\omega_4}{\eta_4 c}\,\chi''_R E_3^2$$

FOUR-WAVE MIXING (HIGH GAIN LIMIT)

FOR $(g_1 + g_3) z >> 1$

$$I_2 \approx \frac{I_{20}}{(1+g_3/g_1)^2} \quad e^{(g_1+g_3)z}$$

$$I_4 \approx \frac{I_{20}}{(1+g_3/g_1)^2} \left(\frac{\omega_4}{\omega_2}\right)^2 \frac{I_3}{I_1} \; e^{(g_1+g_3)z}$$

$$\frac{I_4}{I_2} = \left(\frac{\omega_4}{\omega_2}\right)^2 \frac{I_3}{I_1}$$

FOR GIVEN $g_1 + g_3$, OUTPUT IS MAXIMIZED FOR $g_1 >> g_3$, i.e., WHEN STRONG PUMP IS SEEDED.

AP-I-VG-6639

SOLUTION FOR ARBITRARY NUMBER OF PUMP-STOKES PAIRS, VALID INTO SATURATION IN PHASE-MATCHED SYSTEM:

(J.R. ACKERHALT, PHYS. REV. LETTERS 46, 922 (1981))

$$P_i(\theta) = P_{io}\cos(\beta\Omega_i\theta) - \frac{\omega_{pi}}{\Omega_i} S_{io}\sin(\beta\Omega_i\theta)$$

$$S_i(\theta) = S_{io}\cos(\beta\Omega_i\theta) + \frac{\Omega_i}{\omega_{pi}} P_{io}\sin(\beta\Omega_i\theta)$$

where $\Omega_i = \sqrt{\omega_{pi}\omega_{si}}$, $\beta = \frac{2\pi}{\eta c}\chi_R''$, and

$$\frac{d\theta}{dz} = \left|\sum P_i S_i^*\right|$$

$$\approx \frac{1}{2}\left(\sum I_i\right)\sin\left[2(\beta\langle\Omega\rangle\theta + \langle\xi\rangle)\right],$$

$$I_i = \frac{\Omega_i}{\omega_{pi}}|P_i|^2$$

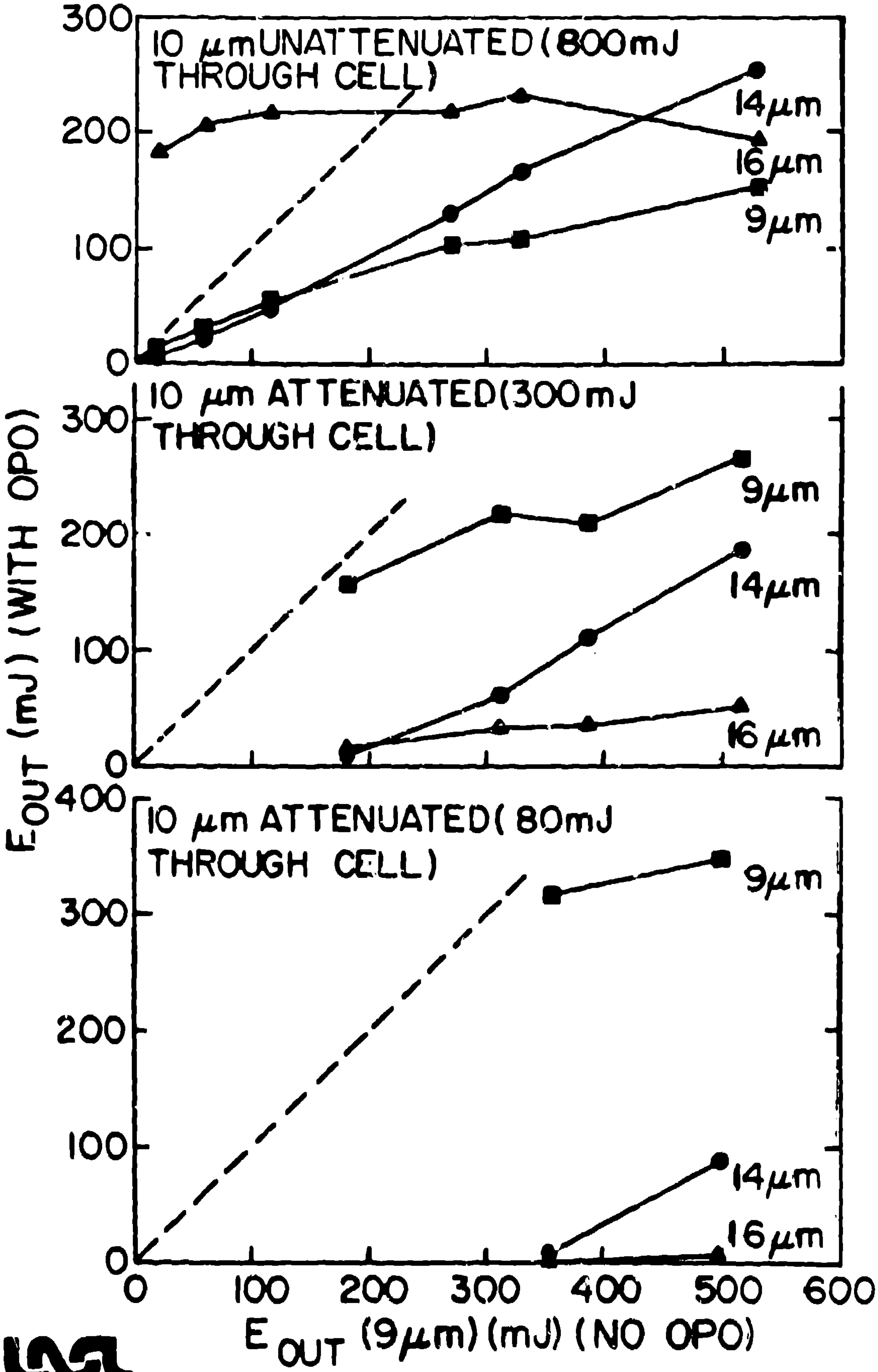
$$\langle\Omega\rangle = \frac{\sum_i I_i\Omega_i}{\sum_i I_i}$$

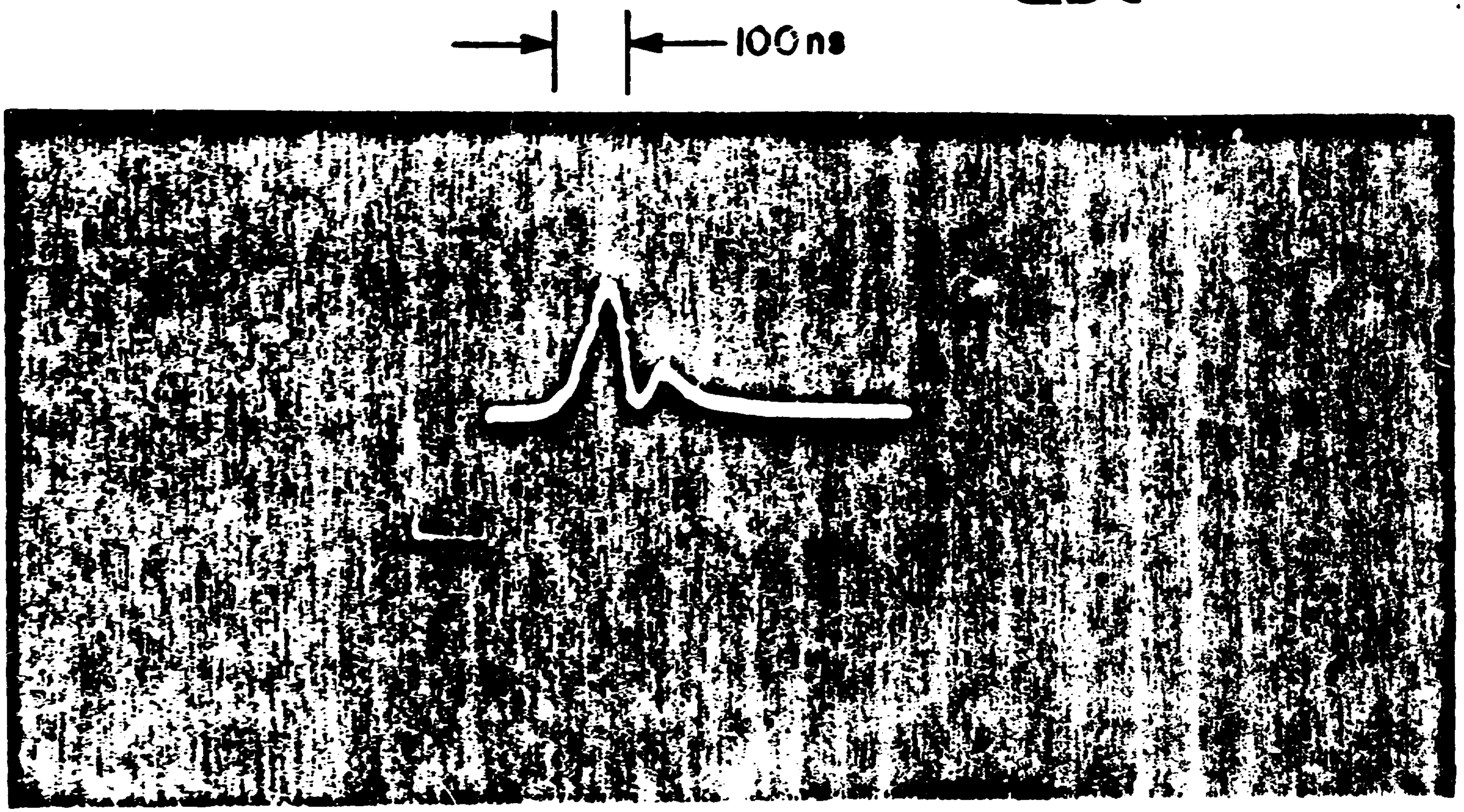
$$\langle\xi\rangle = \frac{\sum_i \mathrm{Re}\, P_{io}S_{io}^*}{\sum_i I_i}$$

$$\tan(\beta\langle\Omega\rangle\theta + \langle\xi\rangle) = \langle\xi\rangle e^{\frac{1}{2}gz}$$

$\sim 1 \Rightarrow$ saturation

where $g = 2\beta\langle\Omega\rangle\sum I_i = 2\beta\sum\frac{\Omega_i^2}{\omega_{pi}}|P_i|^2 = \frac{\pi\chi_R''}{\eta c}\sum\omega_{si}|P_i|^2$


10 μm UNATTENUATED (800mJ THROUGH CELL)
14μm
16μm
9μm
10 μm ATTENUATED (300mJ THROUGH CELL)
9μm
14μm
16 μm
10 μm ATTENUATED (80mJ THROUGH CELL)
9μm
14μm
16μm
E OUT (mJ) (WITH OPO)
E OUT (9μm) (mJ) (NO OPO)
0
100
200
300
400
500
600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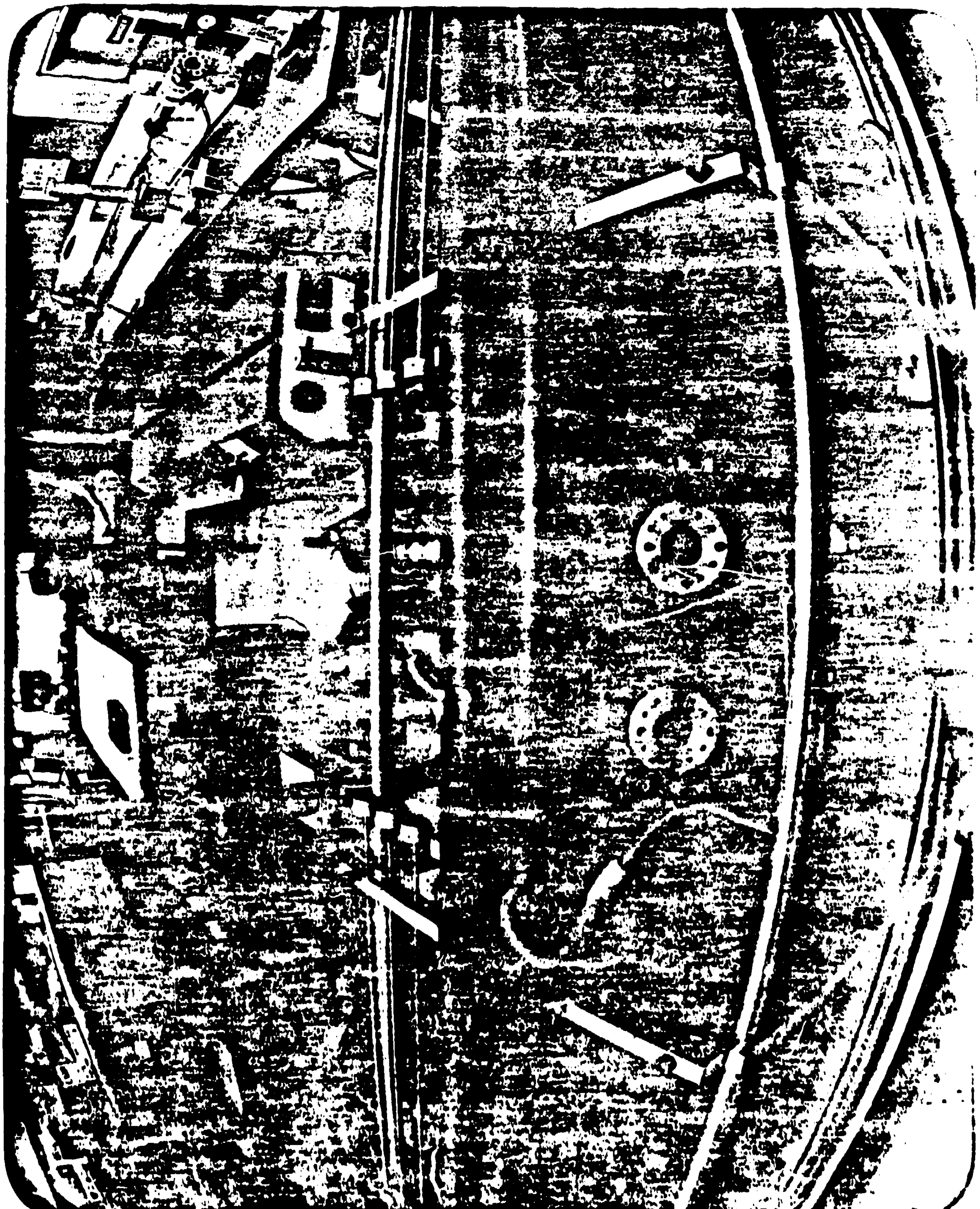

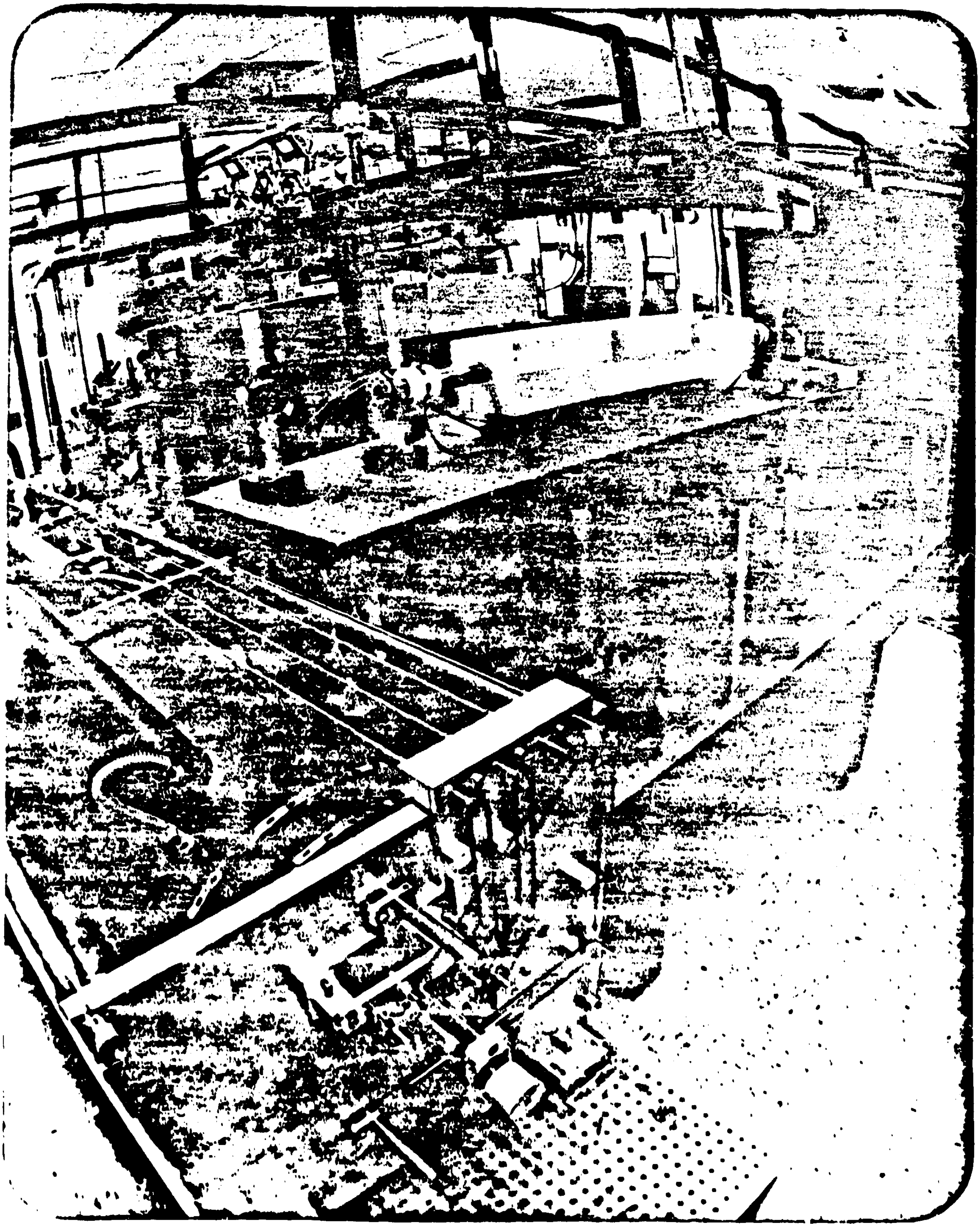

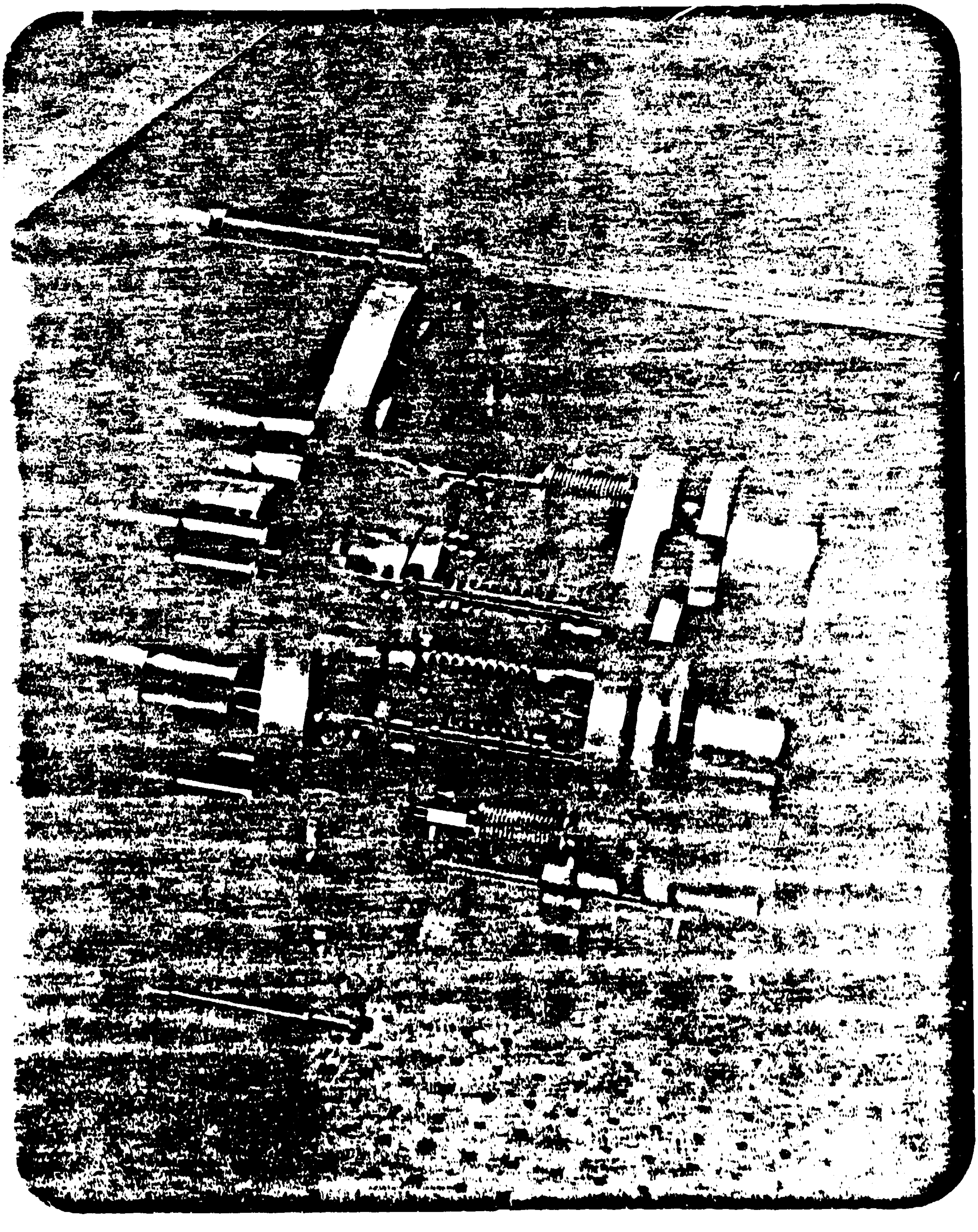

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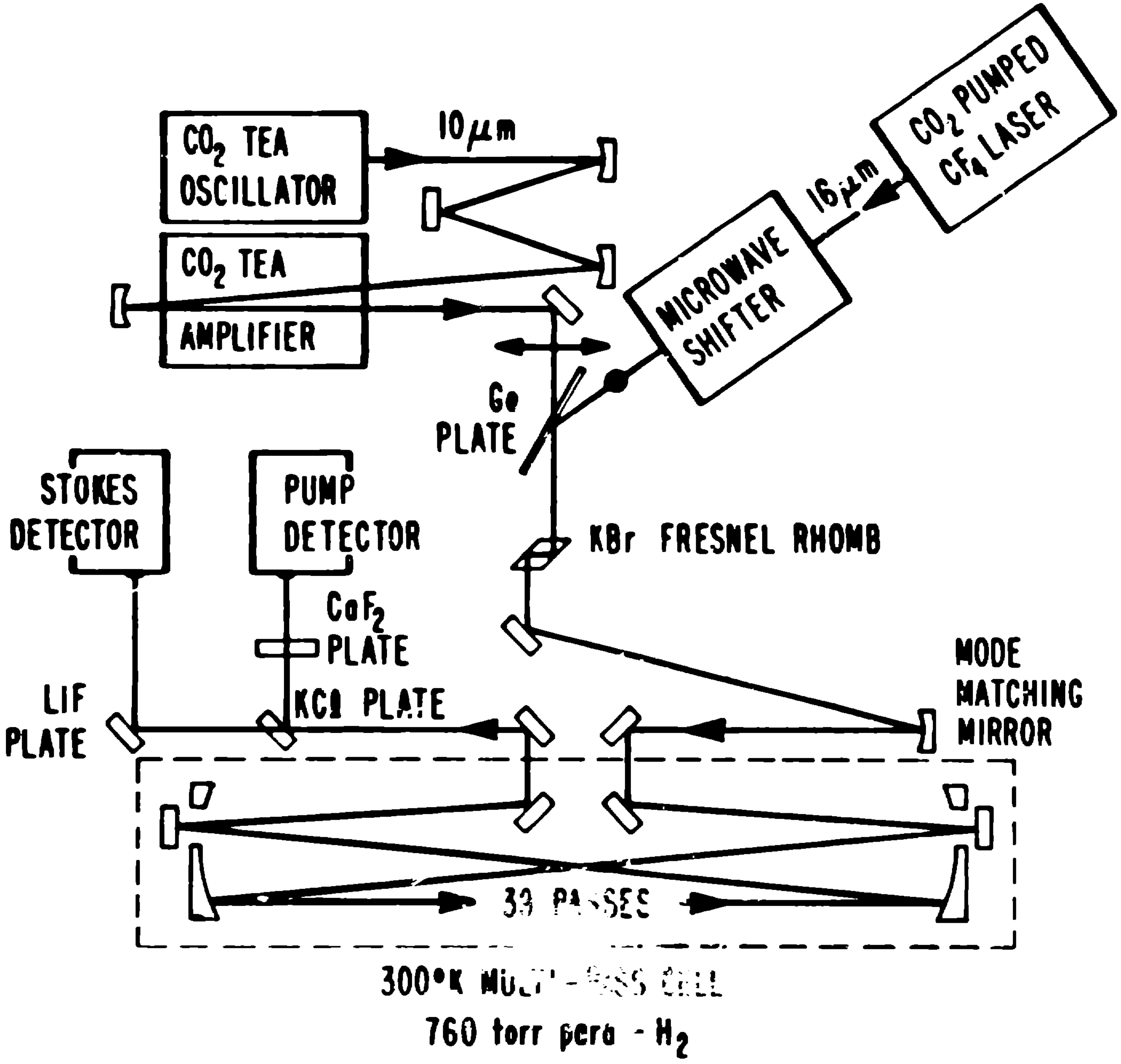
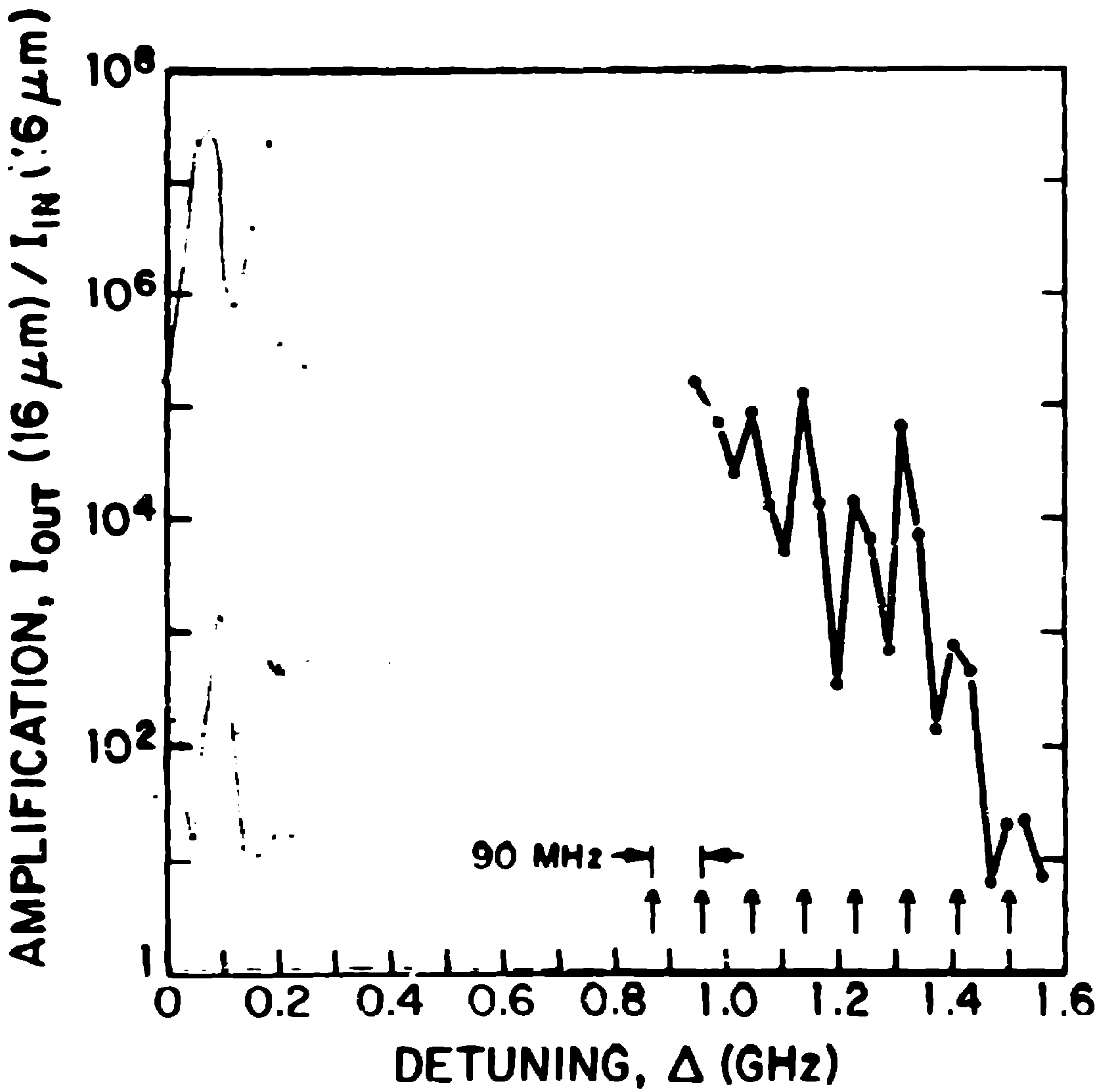
MULTI-PASS CELL APPARATUS

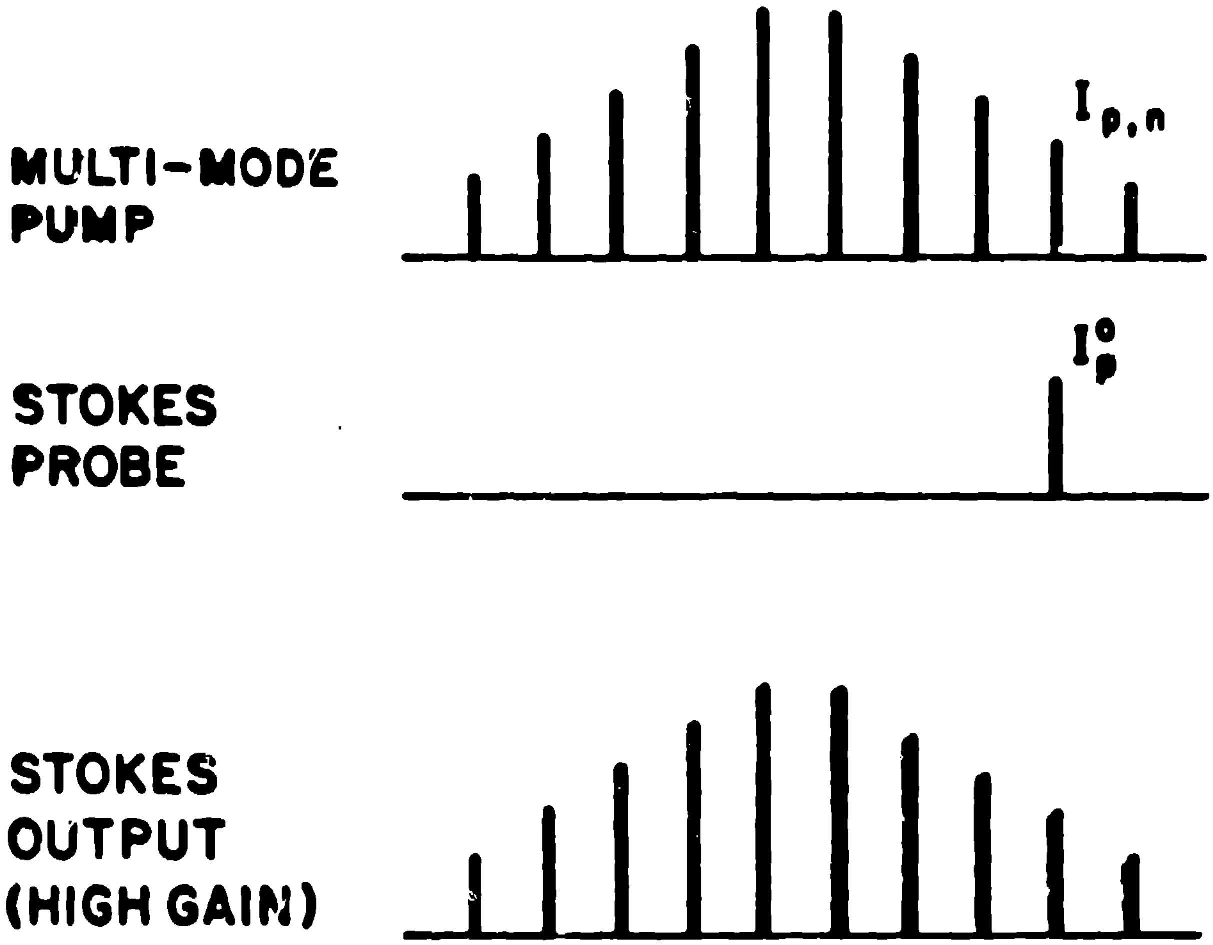


RAMAN AMPLIFIER OF MICROWAVE-SHIFTED CF_4 LASER IN CO_2 PUMPED para-H_2



6471A

RAMAN AMPLIFICATION WITH MULTI-MODE PUMP

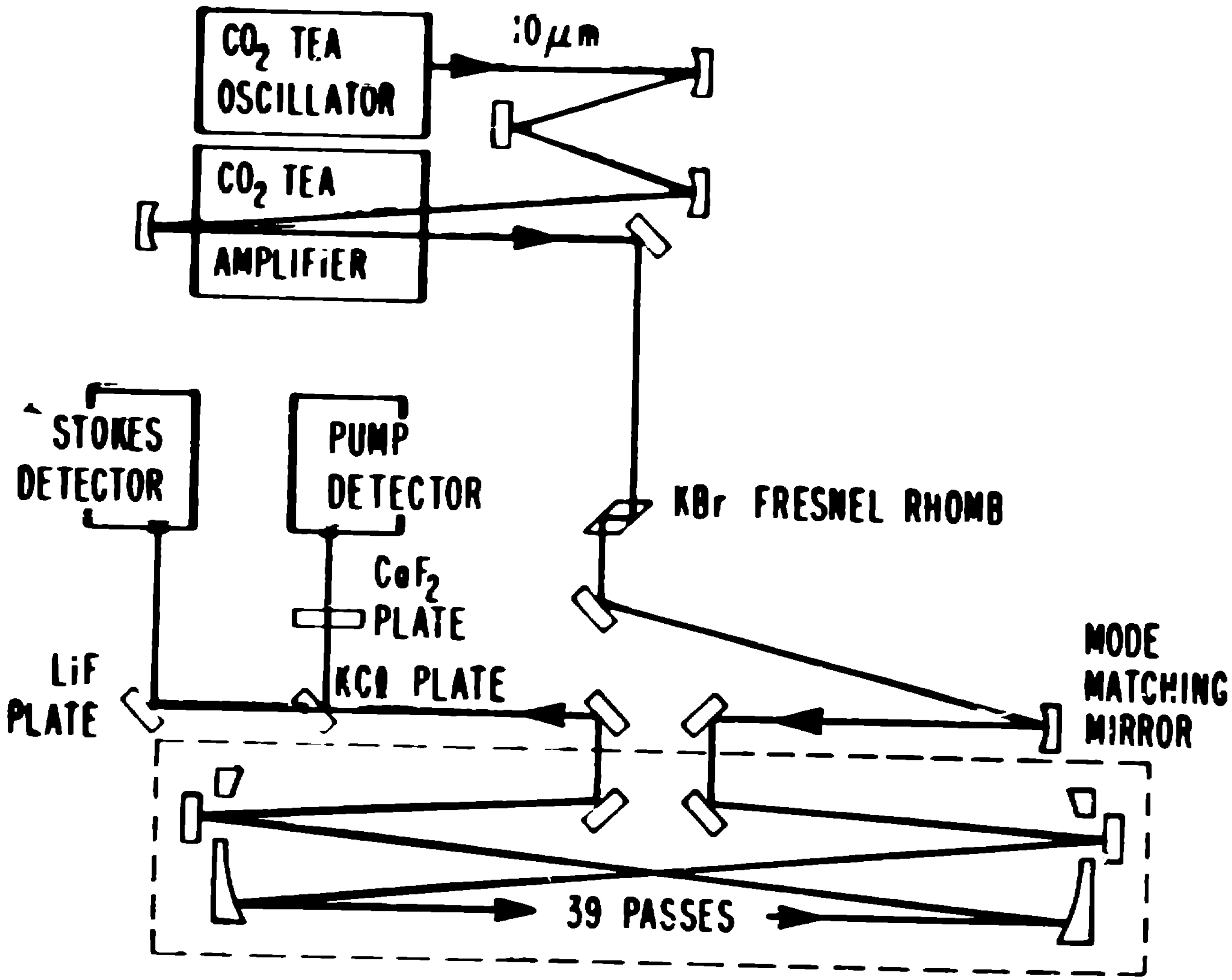


$$\boxed{I_s = I_s^0 \, \frac{I_{p,n}}{I_p} \exp(\alpha I_p z)}$$

$$I_s = \sum_i I_{s,i}$$

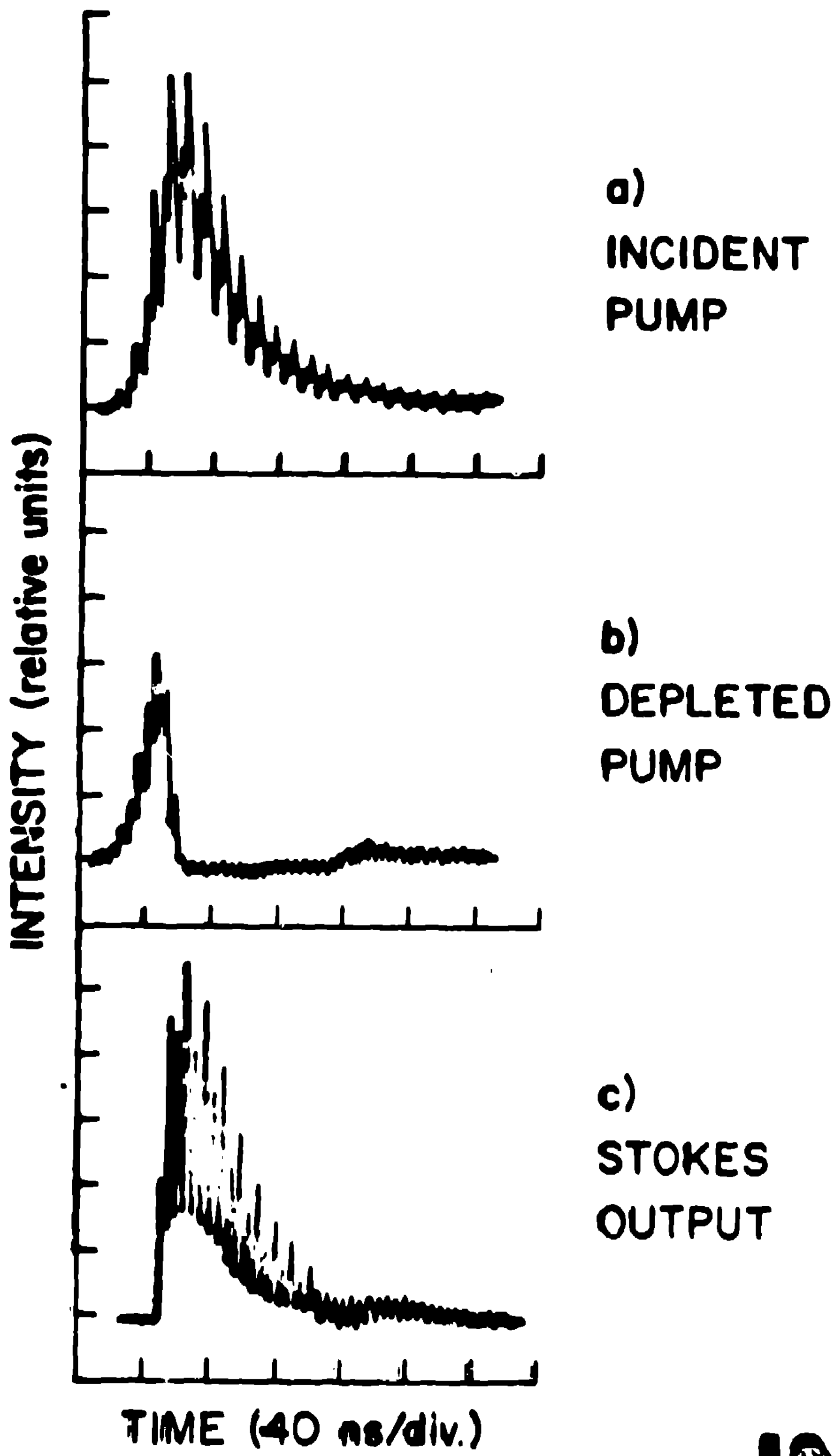
$$I_p = \sum_i I_{p,i}$$

MULTI-PASS CELL APPARATUS

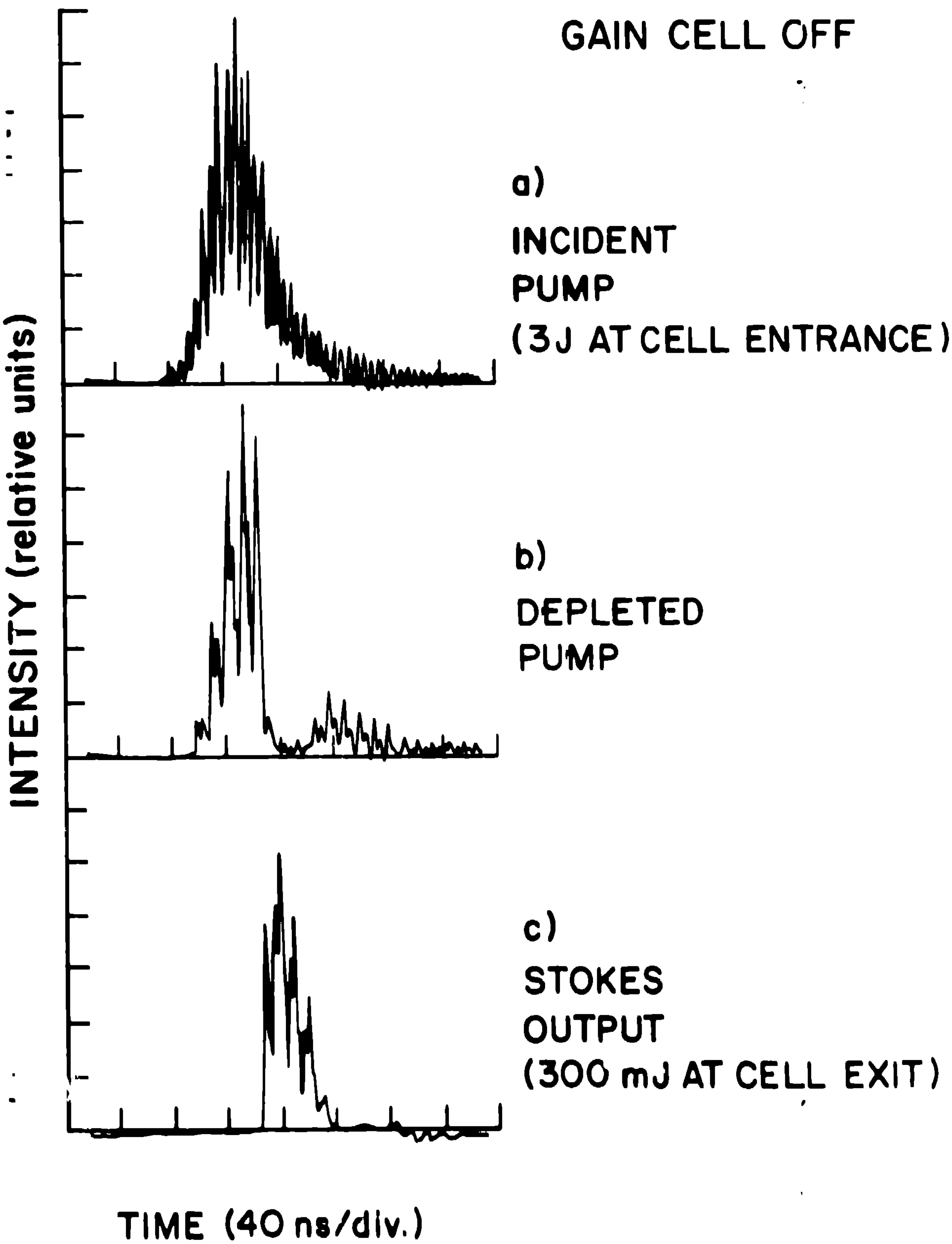


78°K MULTI-PASS CELL
150 torr para - H_2

DEPLETION OF CO_2 PUMP

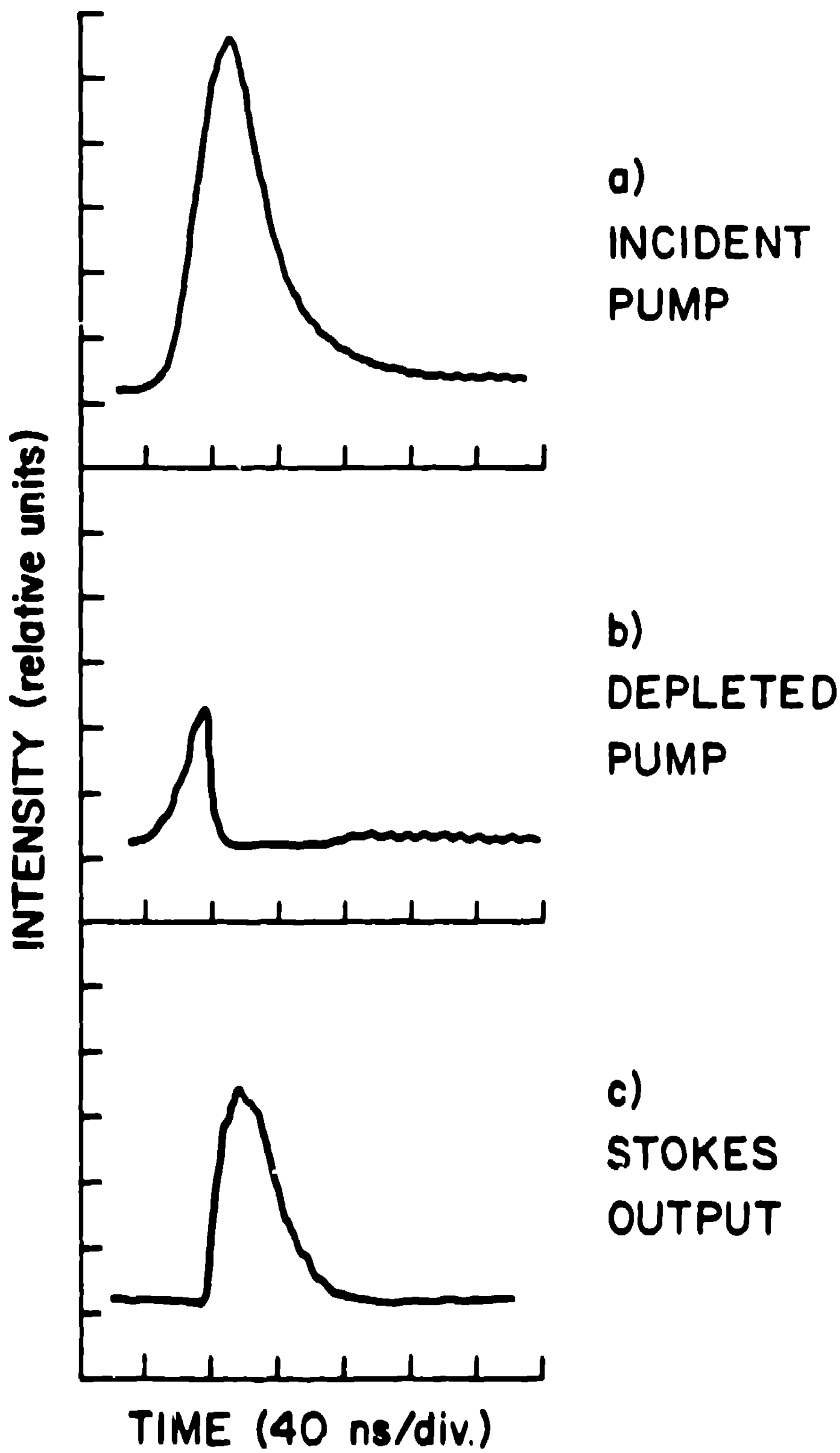


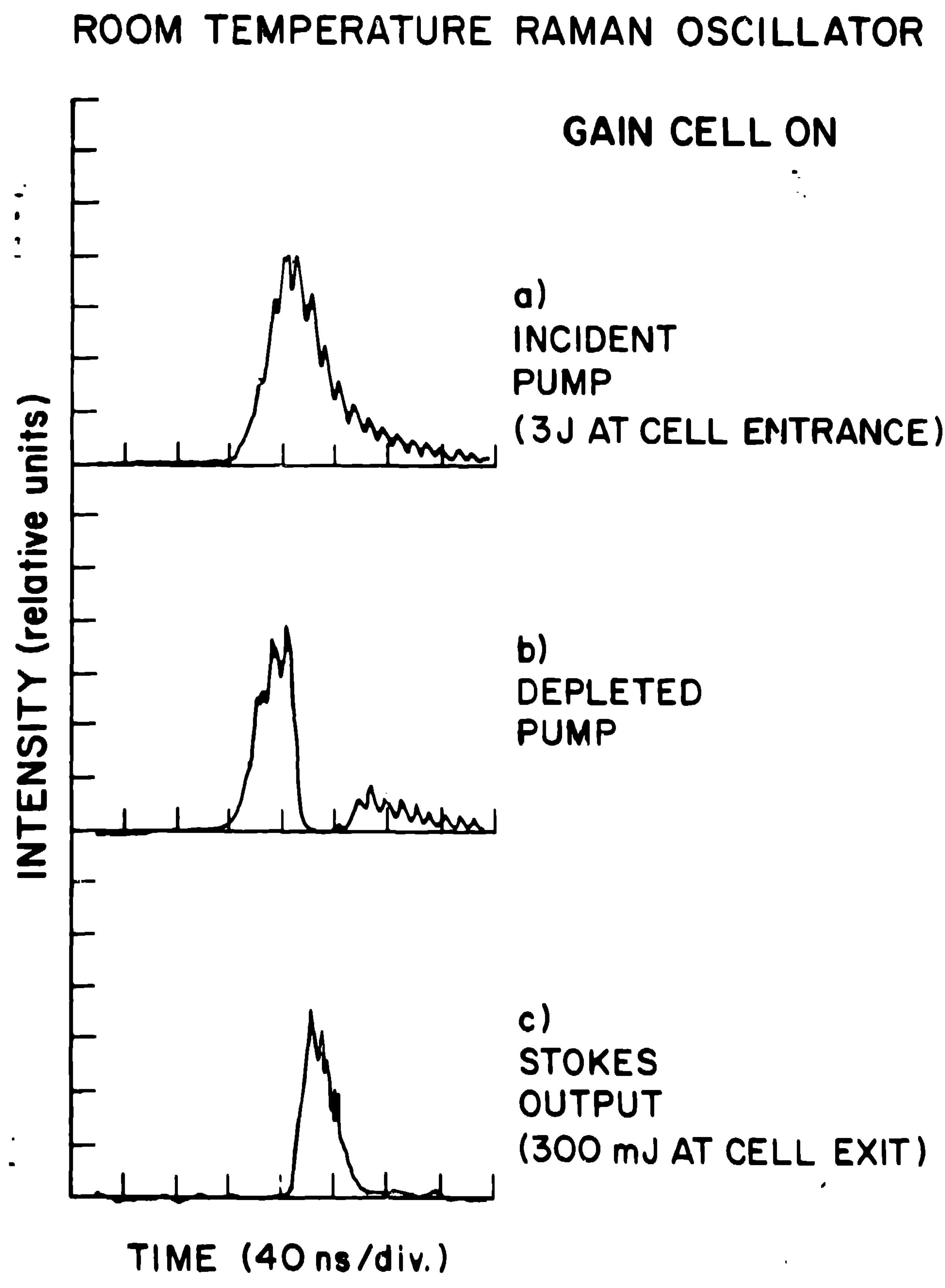
ROOM TEMPERATURE RAMAN OSCILLATOR



AP-I-VG-7231

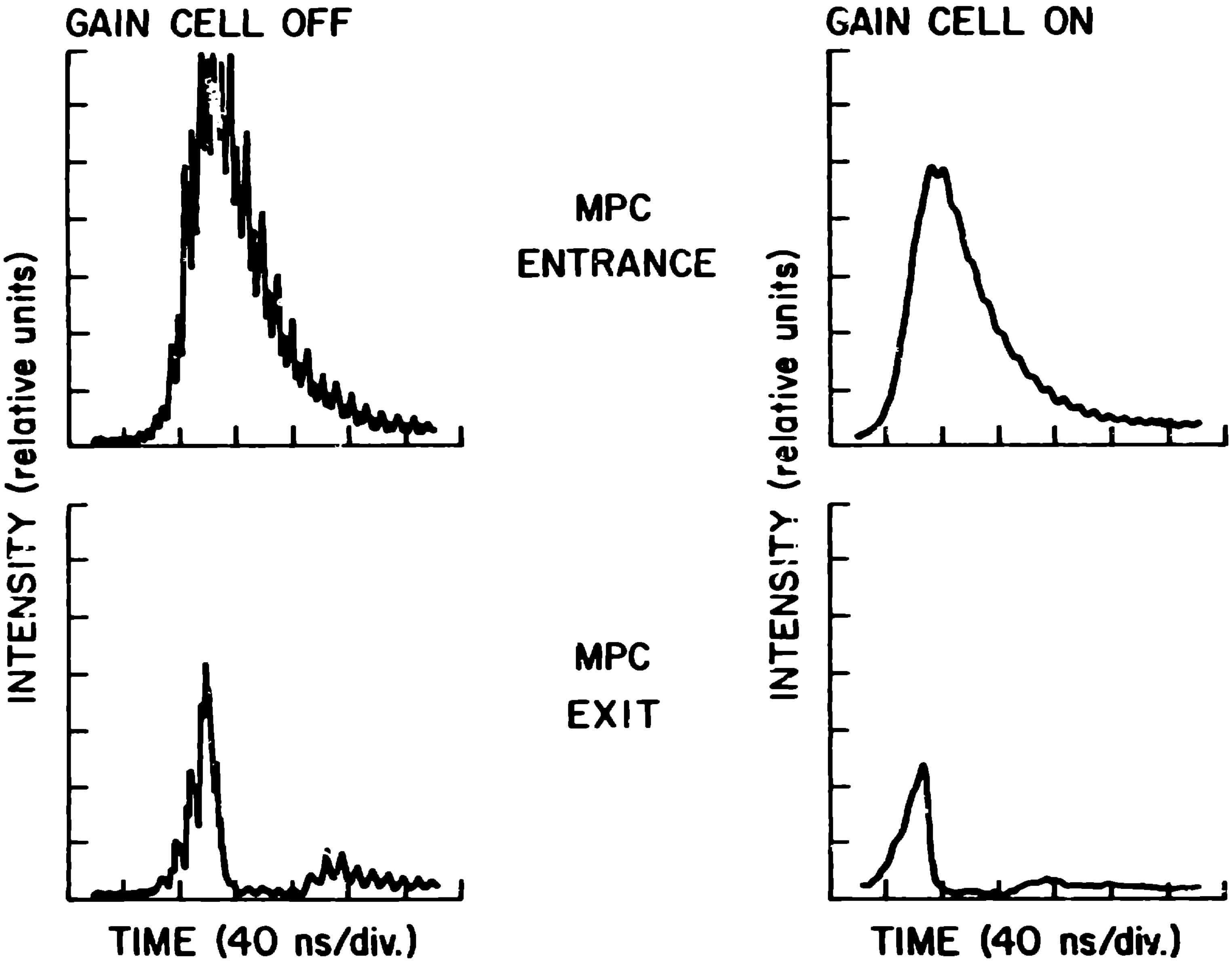
DEPLETION OF CO_2 PUMP




ROOM TEMPERATURE RAMAN OSCILLATOR
GAIN CELL ON
a)
INCIDENT
PUMP
(3J AT CELL ENTRANCE)
b)
DEPLETED
PUMP
c)
STOKES
OUTPUT
(300 mJ AT CELL EXIT)
INTENSITY (relative units)
TIME (40 ns/div.)

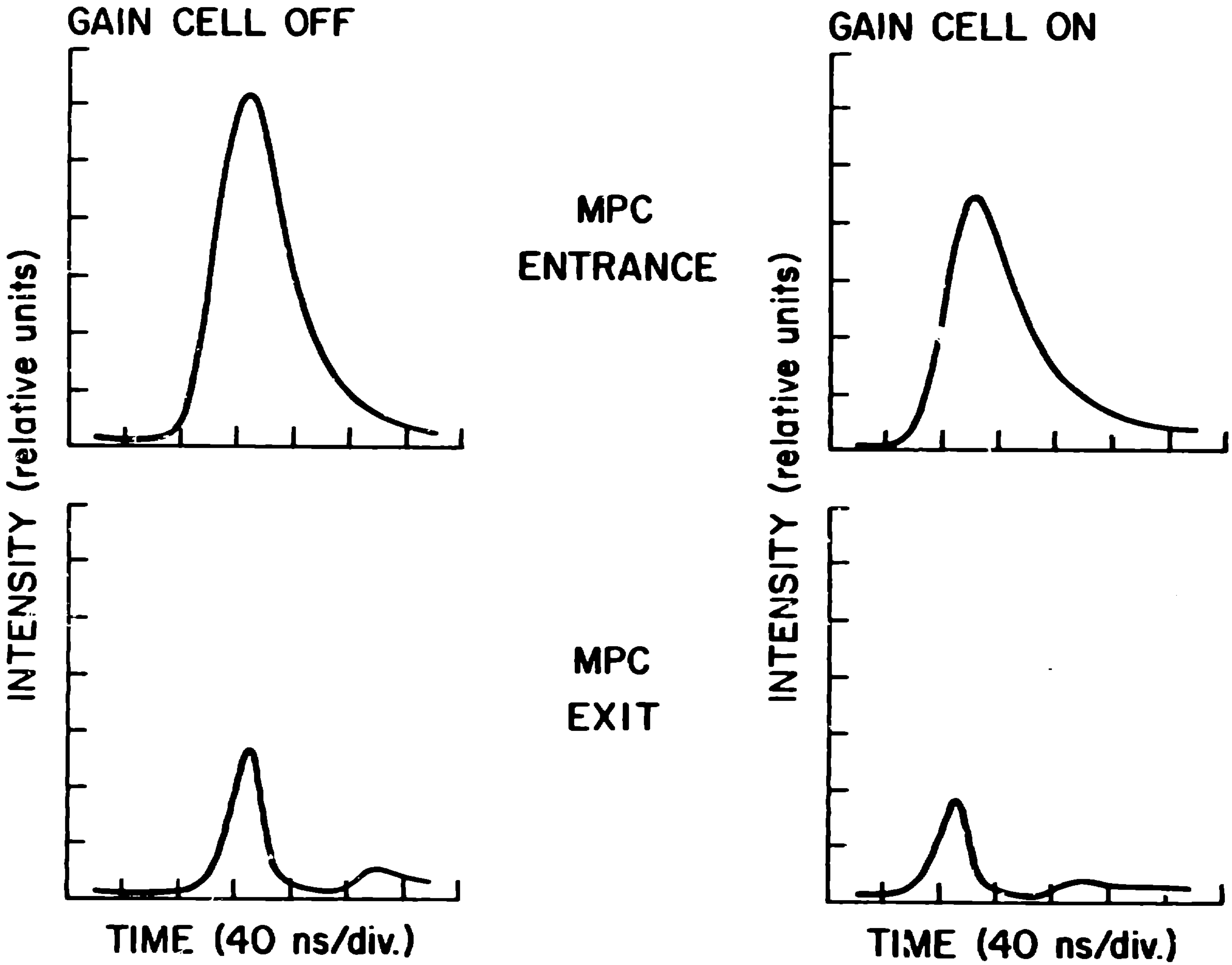
AP-I-VG-7229

CO_2 PUMP DEPLETION
(200 MHz DETECTION)

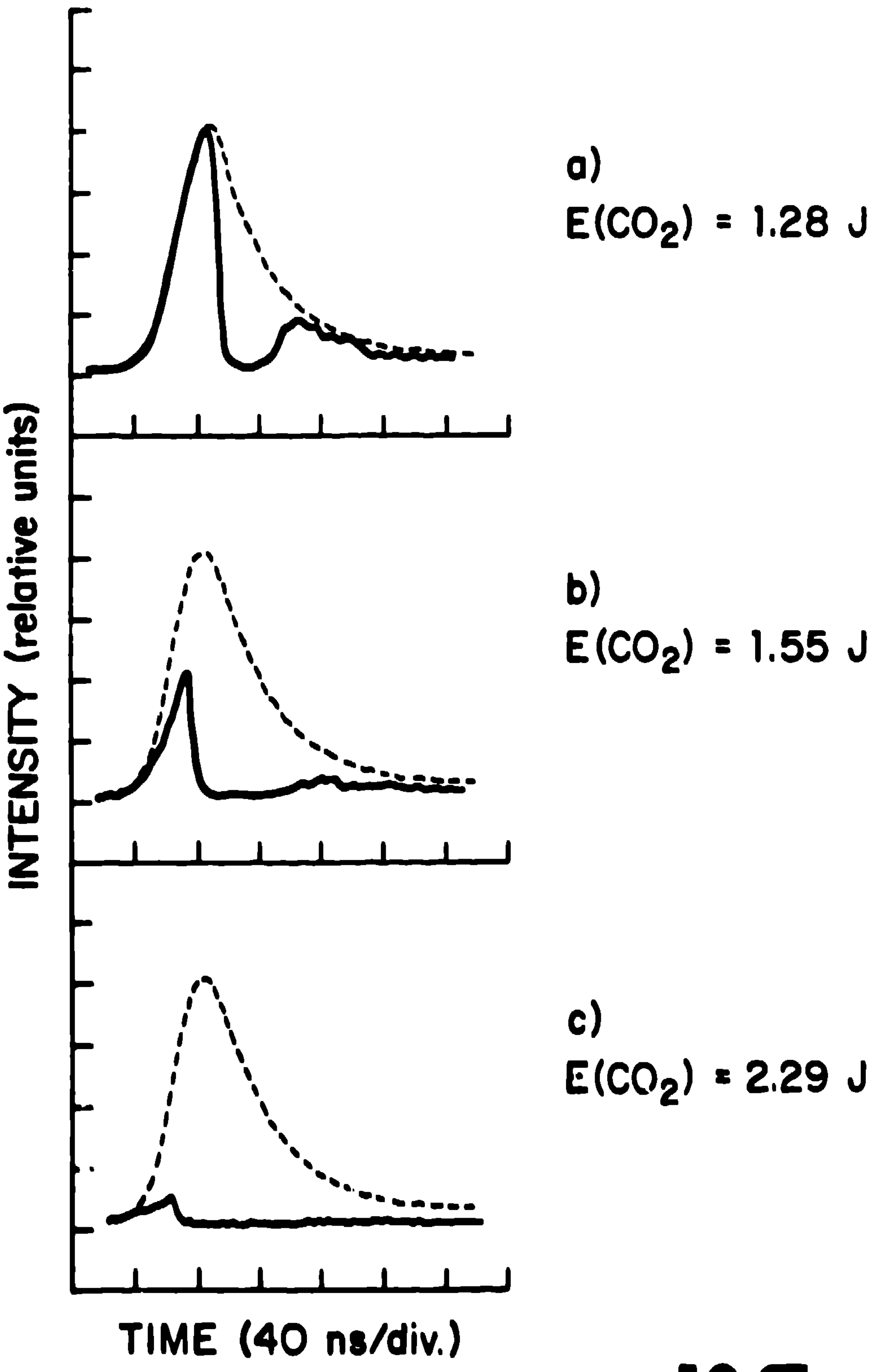


AP-1-VG-6667

<u>CO_2 PUMP DEPLETION</u>
(20 MHz DETECTION)



CO_2 PUMP DEPLETION vs CO_2 PUMP ENERGY



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AP-1-VG-6666