Semi-Annual Report 7

December 1968

STIMULATED RAMAN EMISSION AND ABSORPTION SPECTROSCOPY

B.P. Stoicheff

University of Toronto
Department of Physics

Order No.	NR 015-813/4-14-65	Contract No.	Nonr-5012 (00) M-2
Code No.	5730K21	Expiration Date	31 May 1969
Contractor	The Governors, University of Toronto	Project Scientist	Prof. B.P. Stoicheff
Date of Contract	1 June 1965	Business	(416) 928-2948
		Home	(416) 225-6421

[redacted]

Reproduction in whole or in part is permitted for any purpose of the United States Government.

34

INTENSITY AND GAIN MEASUREMENTS OF THE STIMULATED RAMAN EMISSION IN LIQUID O_2 AND N_2*.

J.B. Grun[†], A.K. McQuillan[‡] and B.P. Stoicheff

Department of Physics, University of Toronto,

Toronto 5, Canada

ABSTRACT

In liquid O_2 and N_2 the threshold for stimulated Raman emission is found to be much lower than for other nonlinear processes. Thus it is possible to make reliable measurements of the intensity of Raman emission over a large range of incident laser power by using a simple longitudinal geometry. Several distinct regions of emission were investigated, including normal Raman scattering, exponential gain, onset of oscillation and saturation. There is good agreement with theory.

INTRODUCTION

It is well-known[1-3] that the comparison of theoretical and experimental values of intensity and gain in stimulated Raman emission is complicated by several competing processes such as self-focusing, and Brillouin and Rayleigh scattering, all of which may have similar appearance thresholds. Thus, anomalous intensity behaviour in many liquids and even in gases[4-6] and solids[7] appears to be the rule rather than the exception. One important consequence is that the premature onset of oscillation has precluded the observation of the expected exponential gain in most materials with the exception of gaseous hydrogen and liquid acetone and carbon tetrachloride[8]. Bloembergen and Lallemand[3,6] have overcome some of these difficulties by the use of a Raman amplifier and have demonstrated its importance in obtaining reliable values of the Raman gain. Other useful experimental arrangements in such studies include, the transverse resonator of Dennis and Tannenwald[9], the off-axis resonator of Jennings and Takuma[10] and the diffusely pumped amplifier of Bortfeld and Sooy[11]. More recently, Shapiro, Giordmaine and Wecht[12], Bret and Weber[13], and Kaiser and Maier[14] have shown that with picosecond and subnanosecond laser pulses stimulated Raman scattering is the dominant nonlinear scattering process in several liquids and thus have obtained good agreement with theoretical intensities.

The present investigation of laser stimulated Raman emission from liquid O_2 and N_2 arose from the results of earlier studies of the spectra of the normal and stimulated scattering. In one, it was shown that the linewidth of the normal Raman scattering was exceptionally narrow indicating a large Raman gain[15]: in another, concerning the stimulated Raman emission, extremely sharp spectral lines were observed (Fig.1) without any evidence of broadening[16], thus indicating that self-focusing and other scattering processes were not prominent. From these results we concluded that possibly the threshold for stimulated Raman scattering is lower than for the competing precesses, in which case liquid O_2 and N_2 would be ideal substances for experimental study. Indeed, the present investigation has shown that liquid O_2 and N_2 are unique in this respect and no self-focusing or stimulated Brillouin scattering has been detected up to the highest incident laser power.

We wish to report our observations of the intensity of Raman Stokes radiation corresponding to the vibrational frequencies 1552.0 cm^{-1} and 2326.5 cm^{-1} of liquid O_2 and N_2 respectively. A simple longitudinal arrangement was used. The range of Raman intensity measurements includes the normal emission which varies linearly with incident laser power, a region of exponential gain over several orders of magnitude, the onset of oscillation with feedback by Rayleigh scattering and finally a region of saturation and depletion. The observed gain is in good agreement with that calculated from our experimentally determined cross-section for scattering.

APPARATUS AND EXPERIMENTAL PROCEDURE

The exciting source was a giant-pulse ruby laser with a rotating prism at one end and at the other a plane parallel reflector (~25% reflectivity) of Corning 2-58 glass which served as a mode selector[16] and also as a filter. The radiation was emitted in a single pulse of ~30 ns duration and in a single (or nearly single) axial mode. Good reproducibility in the laser pulse was obtained by firing the laser at constant power near threshold, at regular (3 min.) intervals with the ruby at a constant temperature (-10°C). This procedure also eliminated any spatial drift of the laser beam at the distant spectrometer slit.

The temporal behaviour of a typical laser pulse is shown in Fig. 2a. A study of the spatial intensity distribution of the laser beam was made at a magnification of 20X and by photographing the beam after attenuation by neutral density filters. This showed the presence of several intensity maxima (Fig.3a) which increased the effective intensity of the laser beam to twice the average intensity. Also, the laser radiation was found to be plane polarized to better than 2000:1.

The longitudinal arrangement shown in Fig.4 was used for the measurements of Raman scattering intensity and state of polarization, in the forward direction. The sample container was a simple dewar of 1 litre capacity with a path length of 5.8 cm between the two inner windows. It was positioned approx. 4 m from the laser in order to reduce possible feedback of scattered radiation to the laser. At each filling of the dewar the liquid was passed through a 5μ millipore filter to remove any dust particles. A short time after a filling, the liquid became quiescent.

In order to increase the laser power density incident on the samples, the beam diameter was reduced by a factor of about 10 (to 0.6 mm) with a system of two lenses. The incident laser power was varied from 30 KW to 600 KW by inserting calibrated neutral density filters of glass in the beam at the entrance diaphragm D_1 and lens L_1. The laser pulse was monitored with an EG & G photodiode (SGD-100) and displayed on a Tektronix 555 (or 519) oscilloscope. An essentially parallel laser beam was incident on the sample. The radiation scattered in the forward direction was collected through the exit diaphragm D_2 and focused on the slit of the spectrometer. The laser light entering the spectrometer was attenuated with calibrated filters. A grating spectrometer (Spex 1700) having a dispersion of 10 Å/mm was used with both entrance and exit slits open to 3 mm. Measurements of Stokes intensities were made with an RCA 7102 Photomultipler having a cooled photocathode (-10°C). The signal was amplified 40 times by a two-stage emitter follower and fed into a type L preamplifier of the oscilloscope. The pulse heights from the oscilloscope traces gave an effective measurement of the intensity of Stokes emission during each laser pulse. Brief studies of the laser and Stokes pulse envelopes were made with a fast photodiode (ITT FW 114A) and a Tektronix 519 oscilloscope. Depolarization measurements were carried out with a Nicol prism placed at the slit of the spectrometer.

Several precautions were taken to reduce any stray light and to minimize its effect on the intensity measurements, especially of the low intensity normal Raman scattering. The main sources of unwanted stray light were found to be the laser flashlamp, and optical filters and lenses of glass along the laser beam which emitted relatively intense fluorescence radiation. Thus, all of the optics and sample dewar were enclosed in a light-tight box having a 6 mm entrance aperture; diaphragms were placed along the laser beam path in front of lenses; and quartz lenses were used instead of glass lenses to minimize the fluorescence. Finally, the effects of the broad band fluorescence were suppressed by the use of a high dispersion spectrometer.

For each liquid, the intensity measurements were carried out in two stages. In the low intensity region of the normal

Raman scattering, the light collecting cone was 1.45×10^{-3} ster. for N_2 and 5.80×10^{-4} ster. for O_2. Calibrated filters were inserted in front of the spectrometer slit to cover the intensity range. In the high intensity region of stimulated Raman emission the light collecting cone was smaller, being 1.30×10^{-4} ster. for both N_2 and O_2. Again, calibrated filters were used to make intensity measurements over approximately ten orders of magnitude. The laser pulse energy was measured with a calibrated thermopile (TRG 100). The many optical filters used to attenuate the laser and Raman radiation were calibrated spectrophotometrically (Beckman DU) each to an accuracy of 3%. The transmission characteristics of the spectrometer and the sensitivity of the photomultiplier were measured over the required wavelength region (and for light of parallel and perpendicular polarization) using a NBS standard lamp.

An estimate of the possible errors in making absolute intensity measurements of the Raman scattering indicated an accuracy of ±50%, the main source of error arising from the many filters used in attenuating the laser radiation. However the accuracy of relative intensity measurements was considered to be better than ±30%.

BRIEF RÉSUMÉ OF THEORY

The theory of stimulated Raman scattering has been developed by many authors, notably, Hellwarth[17], Bloembergen and Shen[18], Townes and co-workers[19] and Maker and Terhune[20]. They have shown that the stimulated Stokes emission grows exponentially from noise according to the relation

$$I_s(\ell) = I_s(0)e^{+g\ell I_o} \tag{1}$$

Here, $I_s(\ell)$ is the intensity of the stimulated Stokes emission, $I_s(0)$ is the intensity of the normal (spontaneous) Stokes emission, I_o is the incident laser power density, and ℓ is the length of the amplifying medium. The gain g is given by

$$g = \frac{2c^2}{\pi h n^2} \cdot \frac{N}{\Delta\nu(\nu_o-\nu_R)^3} \cdot \frac{d\sigma}{d\Omega} \tag{2}$$

In general, and in the present work, g represents the gain for radiation polarized in the same plane as the incident plane-polarized laser radiation. In Eq. (2), c is the velocity of light; h is Planck's constant, n the refractive index, N is the effective number of molecules per cm^3, $\Delta\nu$ is the normal Raman linewidth, $\nu_o-\nu_R$ is the frequency of the Raman line, and $d\sigma/d\Omega$ is the total differential cross-section per molecule per ster. for the one polarization.

The total differential cross-section, $d\sigma/d\Omega$ may be determined from absolute intensity measurements of the normal

Raman scattering. For plane polarized incident light, it is defined as

$$\frac{d\sigma}{d\Omega} = \frac{2^4\pi^4}{c^4}\left[\frac{h}{8\pi^2\nu_R}\right]\frac{d}{\mu}\cdot\frac{(\nu_o-\nu_R)^4}{1-\exp(-h\nu_R/kT)}\cdot K\alpha'^2\left(1+\frac{7}{45}\,\frac{\gamma'^2}{\alpha'^2}\right) \quad (3)$$

according to the polarizability theory of Placzek[21].
Here ν_R is the frequency of the Raman-active molecular vibration, d is the degree of degeneracy of the vibration (=1 for the totally symmetric vibrations), μ is the reduced mass, k and T are the Boltzmann constant and absolute temperature, and α' and γ' are, respectively, the isotropic and anisotropic parts of the derivative of the polarizability with respect to the internuclear coordinate at the equilibrium position. The constant K is the local field correction given by[22]

$$K = \frac{n_s}{n_o}\,(n_s^2+2)^2\,(n_o^2+2)^2/81 \quad (4)$$

where n_o and n_s are the indices of refraction at the laser and Stokes frequencies, respectively. In order to evaluate γ' and α' it is necessary to measure the depolarization ratio $\rho = I_\perp/I_\parallel = 3\gamma'^2/(45\alpha'^2 + 4\gamma'^2)$. Here $I_\perp$ and $I_\parallel$ are the intensities of scattered light polarized $\perp$ and $\parallel$, respectively, to the plane-polarized incident light.

It may be mentioned that Eq.(3) is valid only when the frequency of the incident exciting light is far from the main absorption bands of O_2 and N_2 which occur in the vacuum ultraviolet region.

EXPERIMENTAL RESULTS AND DISCUSSION

The observed intensity of first-order Stokes radiation over a range of incident laser intensity is shown in Fig. 5 for liquid O_2 and in Fig. 6 for liquid N_2. For both liquids, it was possible to investigate the Raman intensity over a range of approx. 12 orders of magnitude, from the very low intensity of normal scattering through a region of exponential amplification and oscillation to an intensity approaching the incident intensity, and finally saturation. These results will be discussed below under the headings (a) normal Raman scattering, (b) exponential gain, and (c) oscillation and saturation.

(a) Normal Raman Scattering.

The region of normal Raman scattering is one of very low intensity. Our measurements for O_2 and N_2 are given in Fig. 7. Although the data show considerable scatter, it is seen that there is a linear dependence of Raman intensity on incident laser intensity, as expected from theory. The slopes of the graphs of Fig. 7 were used to determine values of the differential scattering cross-section.

As already mentioned the errors in making these absolute intensity measurements are approx. ±50% whereas the accuracy of the relative measurements is perhaps ±30%. Thus the present method of determining the absolute Raman intensities was checked by measuring the scattering for the 992cm^{-1} line of liquid benzene and comparing the resultant value of the total differential scattering cross-section dσ/dΩ with values measured by other experimenters. This cross-section is related to experimentally measureable quantities by the equation

$$\frac{d\sigma}{d\Omega} = \frac{P_R}{P_o}\,\frac{1}{N\ell\Omega}$$

Here, P_R is the Raman power for the whole line scattered into the solid angle Ω and P_o is the corresponding laser power, N is the density of molecules per cm^3 in the scattering medium and ℓ is the path length (ℓ= 10 cm in our C_6H_6 experiment). Some of the recent values of dσ/dΩ, for benzene are shown in Table I along with our value of 6.6 ± 3 x 10^{-30} cm^2 per molecule per ster. It is seen that the most accurate values are those obtained by Damen, Leite and Porto[23] with a He-Ne laser and by Skinner and Nilsen[24] with an Ar$^+$ laser, which after correction for the ν^4 frequency dependence, are in very good agreement. We have therefore taken the value dσ/dΩ = 4.50 x 10^{-30} cm^2 for benzene (at λ = 6943Å) as a basis for our evaluations and have measured the ratio of the Raman intensities of the

2326 cm^{-1} line for N_2 and 1552 cm^{-1} line for O_2 relative to the 992 cm^{-1} for C_6H_6.

The results of these intensity measurements are given in Table 11 along with measurements of the depolarization ratio ρ for liquid O_2 and N_2. (We have also included values obtained for liquid CS_2.) The measured values of $d\sigma/d\Omega$ and of ρ were used to calculate values of $\alpha' = d\alpha/dr$, the rate of change of polarizability with nuclear displacement, from Eq. (3) after applying the local field correction K (Eq. (4)) and these are included in Table II. The values $\alpha' = 1.6 \times 10^{-16}$ cm^2 and 1.35×10^{-16} cm^2 for liquid N_2 and O_2, respectively, are the same as the values obtained for gaseous N_2 and O_2 by Stansbury, Crawford and Welsh[25]. Our measured value of ρ for liquid N_2 agrees with that measured in the gas by Cabannes and Rousset[26], but our value of ρ for O_2 is considerably lower than theirs.

(b) Exponential gain.

Under the present experimental conditions, the region of normal Raman scattering appears to hold up to incident laser powers of ~70KW. At higher laser powers, both liquids exhibit regions of exponential gain, as shown by the linear portions of the graphs (plotted on semilog scales) in Figs. 5 and 6. For N_2 this region extends over a range of three orders of magnitude and for O_2, four orders of magnitude of Stokes amplification. These results represent stable regions of gain up to factors of at least e^6 and e^8 for liquid N_2 and O_2 respectively.

Values of the gain, g (exp), were obtained from the slopes of the linear portions of the intensity curves (Figs. 5 and 6). These are given in Table III. Also listed for comparison are calculated values of the gain, g(calc.). The calculated values are based on the scattering cross-section $d\sigma/d\Omega$ evaluated here and on the linewidths 0.067 cm^{-1} for N_2 and 0.117 cm^{-1} for O_2 measured by Clements and Stoicheff[15], making use of Eq. (2). It is seen that the values g(exp) and g(calc) are in good agreement.

(c) Oscillation and Saturation.

For both liquids, the regions of exponential gain are abruptly terminated as shown by the discontinuity in slope of the Stokes intensity curves, Figs. 5 and 6. These sharp changes in slope represent the onset of Raman oscillation with a rapid rise in output power. The oscillation threshold for N_2 occurs at somewhat lower laser power than for O_2, 0.13 MW compared with 0.16 MW for O_2. These values were not significantly affected by tilting the dewar with respect to the incident laser beam or by the prescence of ice particles in the liquids (although in the latter case the experimental error was greatly increased). The onset of oscillation is therefore not considered to arise from reflection at the windows or from scattering by bubbles or dust or ice particles. Also, we have experimentally ruled out the possibility that the rapid rise in output power is caused by self-focusing. Near-field photographs of the laser and Stokes radiation at the exit window show no evidence of filament formation and uniform Stokes emission over the beam cross-section (Fig:3b). No filaments were observed up to the highest laser power used, 1 MW, where the self-focusing length is calculated to be 5 cm for O_2 and 9cm for N_2. The critical power for self-focusing[27, 28] calculated from the known Kerr constants[29] is 200KW for O_2 and 600KW for N_2. The observed onset of oscillation occurs at lower laser powers as mentioned above. Moreover, the ratio of laser power at threshold of oscillation in O_2 to that in N_2 was measured to be 1.20±0.006 as compared with the ratio of 0.3 for the respective critical powers for self-focusing.

We believe that the most likely cause of oscillation is feedback of Stokes radiation scattered in the backward direction by Rayleigh scattering. This is suggested by the high Raman gain for these liquids and by the ratio of 1.2 for the gain constants of O_2 and N_2, which is the same value as the ratio of laser power for oscillation. The Rayleigh scattering intensity determined by Stansbury, Crawford and Welsh,[25] for gaseous O_2 and N_2 together with the local field factor for the liquids leads to a feedback factor of $\rho d\sigma/d\Omega$ (Rayleigh) = 6.1×10^{-6} per cm per unit solid angle for liquid N_2 and 6.6×10^{-6} units for O_2. For the effective solid angle of our experiments ($\sim 10^{-4}$ ster.) the feedback factor is approx. 10^{-9} per cm, which is sufficient to explain the onset of oscillation.

In the region of oscillation the rise in Stokes intensity is very steep and represents an increase of five orders of magnitude for liquid O_2 and seven orders of magnitude for liquid N_2.[6,30] The uppermost portions of the intensity curves (Figs. 5 and 6) are similar and indicate strong depletion of laser radiation and conversion to Stokes radiation. The oscilloscope trace in Fig. 2b shows a typical pulse of Stokes radiation in this region with the corresponding laser pulse severely distorted. This process results in the flat tops of the intensity curves and is the region of saturation. At still higher incident laser powers, the first-order Stokes radiation is converted to second-order Stokes (and anti-Stokes radiation) which results in depletion of the first-order Stokes intensity. This depletion is shown in the oscilloscope trace of Fig. 2c.

A brief study of the conversion of laser radiation to first-order Stokes radiation for liquid N_2 was carried out and the results are presented in Fig. 8. Here is plotted the ratio P_{out}/P_{in}, normalized for the laser radiation. The general behaviour of this ratio is in good agreement with the theory of Shen and Bloembergen[18]. Fig. 8 shows high conversion of approx. 75% laser radiation to first-order Stokes radiation in the saturation region.

CONCLUSION

This experiment has shown that liquid N_2 and O_2 are important materials for the study of stimulated Raman scattering. Because of their high Raman gain the stimulated Raman effect emerges as the dominant nonlinear process in these liquids. Thus it was possible to investigate the intensity characteristics and build-up of Stokes radiation over a range of 12 orders of magnitude, from the low intensity normal scattering through exponential amplification, oscillation and saturation and eventual depletion. A detailed study of the region of normal scattering and exponential gain shows very good agreement with theory. The regions of higher intensity also reveal the expected theoretical behaviour and warrant closer study. Finally, the high conversion efficiency of laser to Raman Stokes radiation indicates that these liquids are very useful as new frequency sources.

We are very grateful to Dr. Fujio Shimizu for many helpful discussions.

Footnotes and References

*This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Projects Agency, the U.S. Office of Naval Research, and the Department of Defense. Also supported by the National Research Council, Canada, and the University of Toronto.

†On leave from Laboratoire de Spectroscopie, Université de Strasbourg, France.

‡Holder of Province of Ontario Government Scholarships 1965-68.

1. J.F. McClung, W.G. Wagner and D. Weiner, Phys. Rev. Letters 15, 96 (1965): in Physics of Quantum Electronics, P.L. Kelley, B. Lax P.E. Tannenwald, Eds (McGraw Hill, New York, 1966) pp. 155-158.
2. G. Bret, Compt. Rend. 259, 2991 (1964); 260, 6323 (1965): G. Bret and G. Mayer, in Physics of Quantum Electronics (1966) pp. 180-191: G. Bisson, G. Bret, M. Denariez, F. Gires, G. Mayer, M. Paillette, J. Chimie Phys. (Paris) 64. 197 (1967)
3. P. Lallemand and N. Bloembergen, Appl. Phys. Letters. 6, 210, 212 (1965): in Physics of Quantum Electronics (1966) pp. 137-154.
4. R.W. Minck, E.E. Hagenlocker and W.G. Rado, Phys. Rev. Letters 17, 229(1966)
5. G. Bret and M. Denariez, Phys. Letters, 22, 583 (1966).

6. N. Bloembergen, G. Bret, P. Lallemand, A. Pine and P. Simova, IEEE J. Quant. Electr. QE3, 197 (1967).

7. G. Bisson and G. Mayer, J. de Phys. 29, 97 (1968).

8. G. Bret and M. Denariez, J. Chimie Phys. (Paris) 64, 222 (1967).

9. J.H. Dennis and P.E. Tannenwald, Appl. Phys. Letters 5, 58 (1964).

10. D.A. Jennings and H. Takuma. Appl. Phys. Letters 5, 239 (1964)

11. D.P. Bortfeld and W.R. Sooy, Appl. Phys. Letters, 7, 283 (1965)

12. S.L. Shapiro, J.A. Giordmaine and K.W. Wecht, Phys. Rev. Letters, 19, 1093 (1967).

13. G. Bret and H.P. Weber, IEEE J. Quant. Electronics QE-4, 28 (1968).

14. W. Kaiser and M. Maier, IEEE J. Quant. Electronics QE-4, 67 (1968); private communication 1968

15. W.R.L. Clements and B.P. Stoicheff, Appl. Phys. Letters, 12, 246 (1968).

16. B.P. Stoicheff, Phys. Letters, 7, 186 (1963); Proceedings Enrico Fermi International School of Phys, Course 31, P.A. Miles, Ed (Academic Press, New York 1964) pp. 306-325.

17. R. Hellwarth, Phys. Rev. 130, 1850 (1963); Current Sci (India) 3. 129 (1964).

18. N. Bloembergen and Y.R. Shen Phys. Rev. 133A 37 (1964); 137, A 1787 (1965).

19. E. Garmire, E. Pandarese and C.H. Townes, Phys. Rev. Letters, 11, 160 (1963); R.Y. Chiao, E. Garmire and C.H. Townes, Proceedings Enrico Fermi International School of Phys. Course 31, P.A. Miles, Ed. (Academic Press, New York 1964) pp. 326-338.

20. P.D. Maker and R.W. Terhune, Phys. Rev. 137, A801 (1965).

21. G. Placzek in *Handbuch der Radiologie* (E. Marx, Ed, Akademische Velagsges, Leipzig 1934) 6, p.205.

22. G. Eckhardt and W.G. Wagner, J. Mol. Spectroscopy 19, 407 (1966)

23. T.C. Damen, R.C.C. Leite and S.P.S. Porto, Phys. Rev. Letters, 14, 9 (1965).

24. J.G. Skinner and W.G. Nilsen, J. Opt. Soc. Am. 58, 113 (1968).

25. E.J. Stansbury, M.F. Crawford and H.L. Welsh, Can. J. Phys. 31, 954 (1953).

26. J. Cabannes and A. Rousset, Compt Rend. (Paris) 206, 85 (1938).

27. R.Y. Chiao, E. Garmire and C.H. Townes, Phys. Rev. Letters 13 479 (1964).

28. P.L. Kelley, Phys. Rev. Letters. 15, 1005 (1965).

29. R. Guillien, Physica 3, 895 (1963).

30. cf. P.V. Avizonis, K.C. Jungling, A.H. Guenther, R.M. Heimlich and A.J. Glass, J. Appl. Phys. 39, 1752 (1968).

Table I

Values of the total differential scattering cross-section for the 992 cm^{-1} Raman radiation of liquid benzene.

Authors	$(d\sigma/d\Omega)\times10^{-30}$ $cm^{2}sr^{-1}$	$(d\sigma/d\Omega)\times10^{-30}$ corrected for 6943Å
Damen, Leite, Porto[23]	6.7±1.2 (6328Å)	4.5[a]
Skinner and Nilsen[24]	37.5±4 (4880Å)	4.95[b]
McClung and Weiner[22]	5.9±3 (6943Å)	5.9
Bret et al[2]	9 (6943Å)	9
Present authors	6.6 ±3 (6943Å)	6.6

[a] Calculated from $I\alpha(\nu-\nu_v)^4$, since the frequency of the exciting radiation, ν, is far from principal absorption frequencies. The Raman vibrational frequency is ν_v.

[b] Calculated from $I\alpha(\nu-\nu_v)^4/(\nu_a-\nu)^2$, since the frequency of the exciting radiation is near an absorption frequency, ν_a.

Table II

Values of the total differential cross-section, derivative of the polarizability and depolarization ratio.

Liquid	ν_v cm^{-1}	$\frac{(d\sigma/d\Omega)\ \text{Liquid}}{(d\sigma/d\Omega)\ C_6H_6}$	$(d\sigma/d\Omega)\times10^{30}$ cm^2 sr^{-1}	$\alpha' \times 10^{16}$ cm^2	ρ
O_2	1552.0	0.056±0.017	0.250±0.075	1.35 (1.4)[6]	0.11±0.01
N_2	2326.5	0.041±0.012	0.185±0.055	1.60 (1.6)[b]	0.10±0.01
C_6H_6	992.2	1.00	4.5[a]	2.84	-
CS_2	655.6	2.03±0.60	9.1±2.7	2.91	0.17±0.02

[a] Value of Damen, Leite, Porto[23] corrected for 6943Å radiation

[b] Values given by Stansbury, Crawford, Welsh[25] for gaseous O_2, N_2

Table III.

Values of the Raman gain

Liquid	$(\rho d\sigma/d\Omega)10^8$ $cm^{-1}sr^{-1}$	$\Delta\nu^a$ cm^{-1}	$g(calc)10^2$ $cmMW^{-1}$	$g(exp)10^2$ $cmMW^{-1}$
O_2	0.45±0.14	0.117	1.45±0.4	1.60±0.50
N_2	0.29±0.09	0.067	1.70±0.5	1.60±0.55
C_6H_6	3.06	2.15	0.28	-
CS_2	7.55	0.50	2.4	-

[a] Values of linewidths measured by Clements and Stoicheff.[15]

Figure Captions

Fig. 1. Stimulated Raman spectra of liquid O_2 and N_2 showing the first-order Stokes vibrational lines at 1552.0 and 2326.5 cm^{-1} respectively. The resolving power of the grating spectrograph is 10^5.

Fig. 2. (a) Typical laser pulse monitored with an ITT FW114A photodiode and displayed on a Tektronix 519 oscilloscope. (b) Typical first-order Stokes pulse obtained in the saturation region, and the corresponding depleted laser pulse at the right. (c) Same as (b) but with the Stokes pulse also showing some depletion at higher laser power.

Fig. 3. Near-field patterns showing the spatial intensity distribution of the incident laser beam (a) and the first Stokes emission (b), magnified 20X. Mottled appearance of Stokes picture caused by laser attenuating filters.

Fig. 4. Diagram of apparatus used for Raman intensity measurements. Explanation of symbols D-diaphram, A.F. - attenuating filter, G.G.-ground glass, P.D. - E.G. & G photodiode (SGD-100), L-lens, F-filter.

Fig. 5. Experimental curve for liquid oxygen showing Raman Stokes power as a function of incident ruby laser power.

Fig. 6. Experimental curve for liquid nitrogen showing Raman Stokes output power as a function of incident ruby laser power.

Fig. 7. Experimental measurements of normal Raman scattering for liquids, benzene, oxygen and nitrogen. (Experimental scatter results from very low light levels used, necessitating a high amplification of the photomultiplier signal.)

Fig. 8. Experimental curves showing how the ratios of the laser power (P_L) and the first Stokes power (P_s) at the exit of the dewar, to the incident laser power (P_{Lo}), vary with the incident laser power. The dashed curve in the depleted laser region is only approximate as the laser pulse was severely distorted.

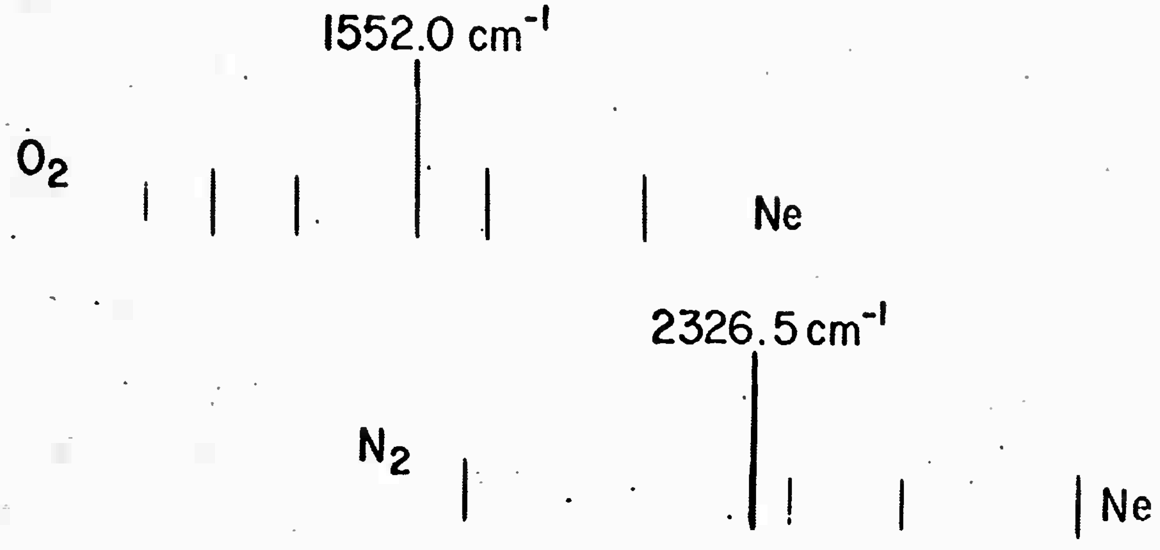


FIG. 1.

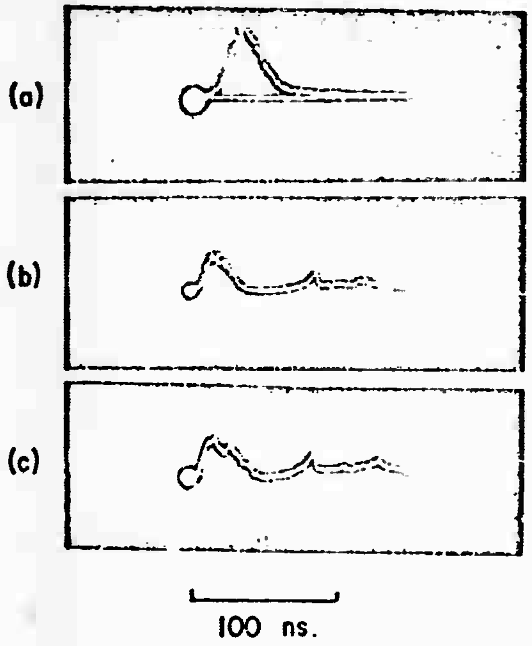


FIG. 2.

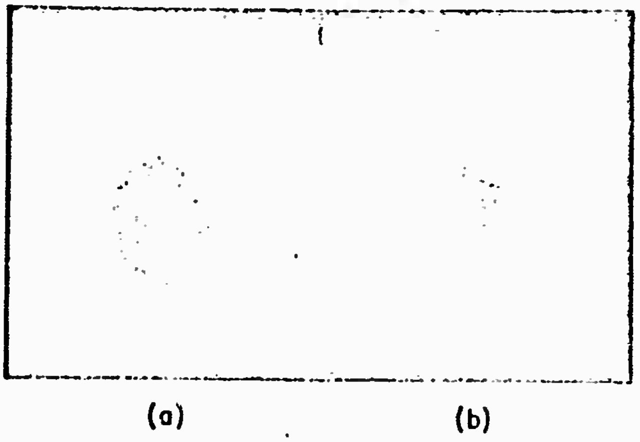
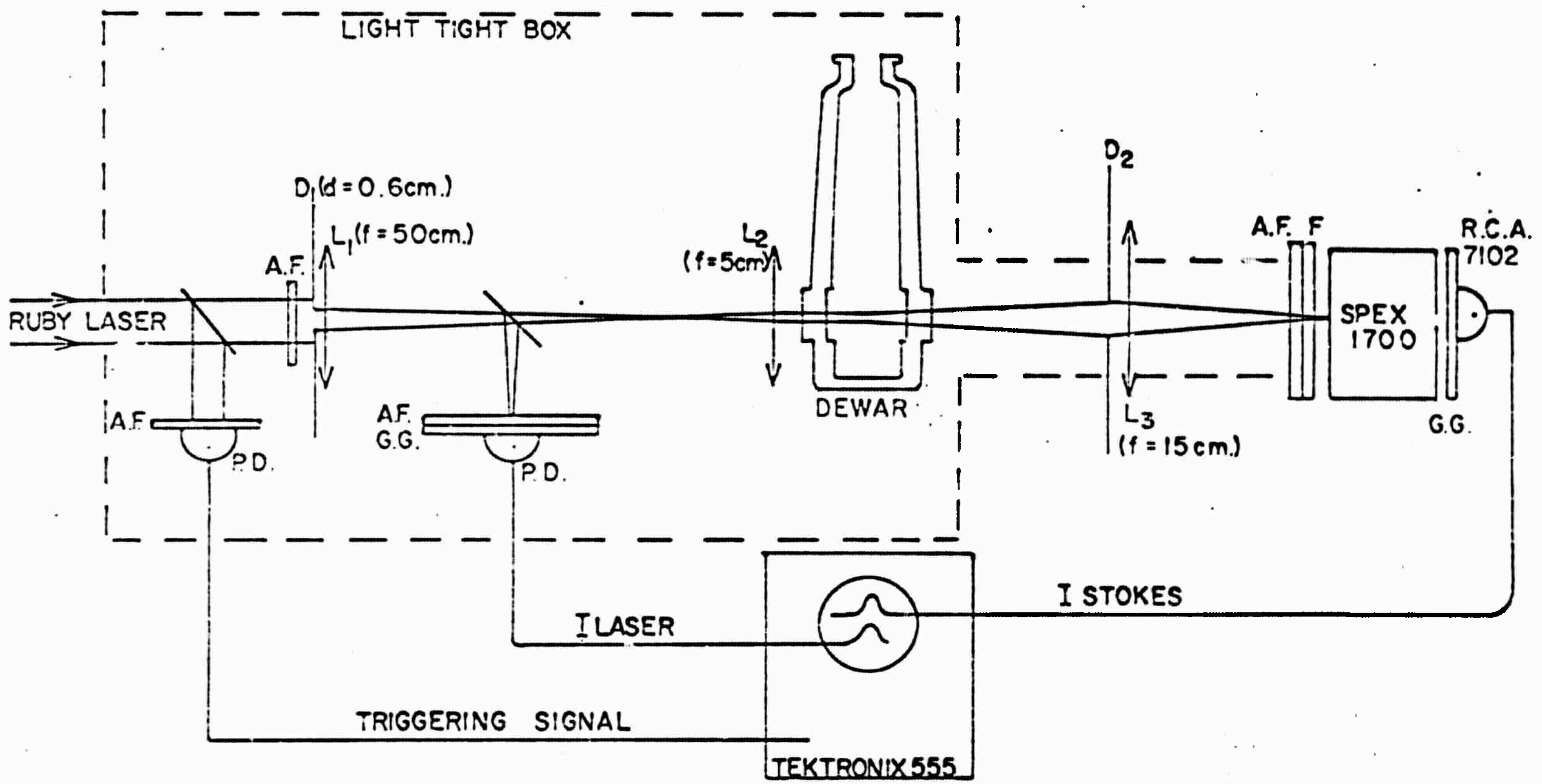
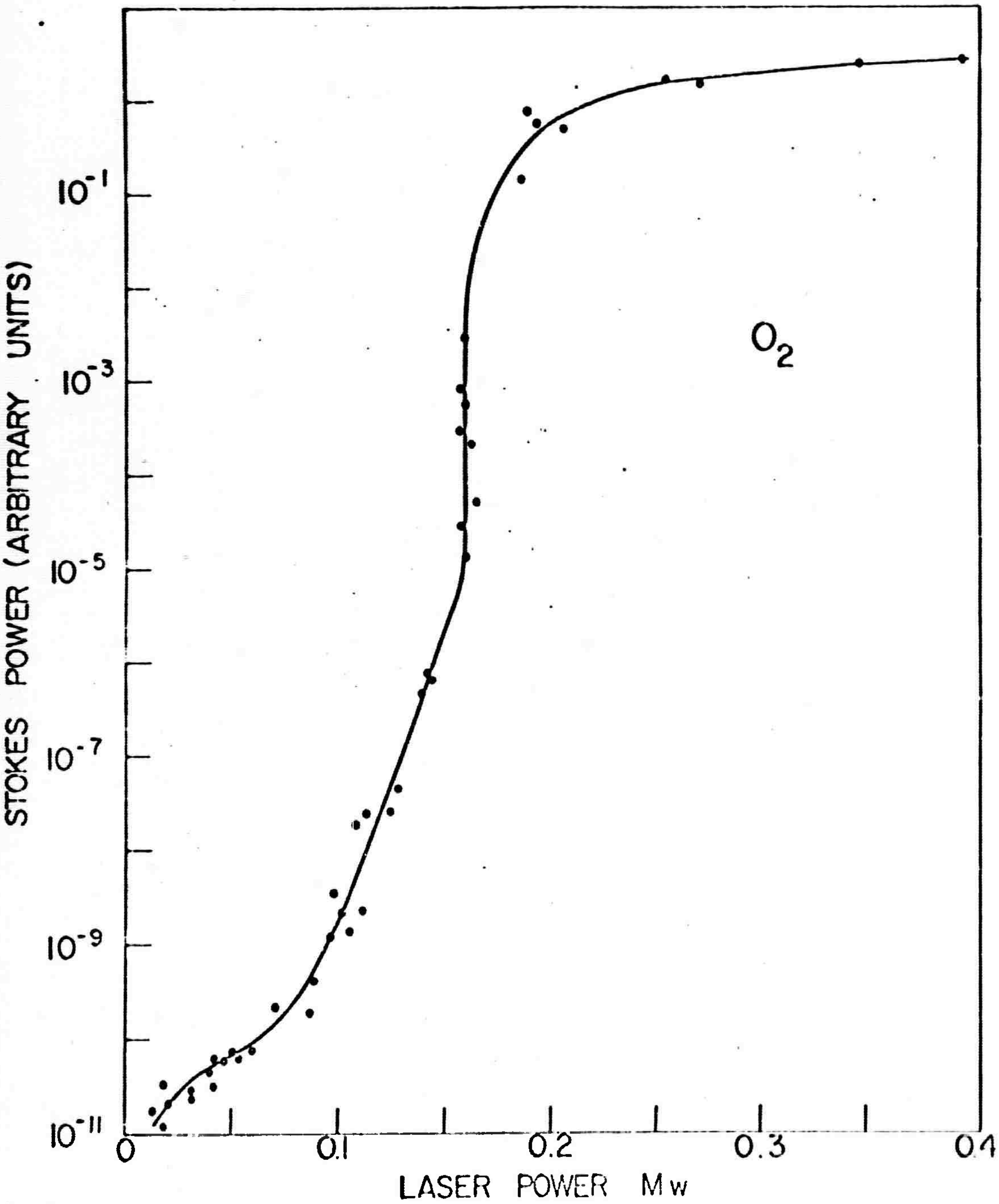


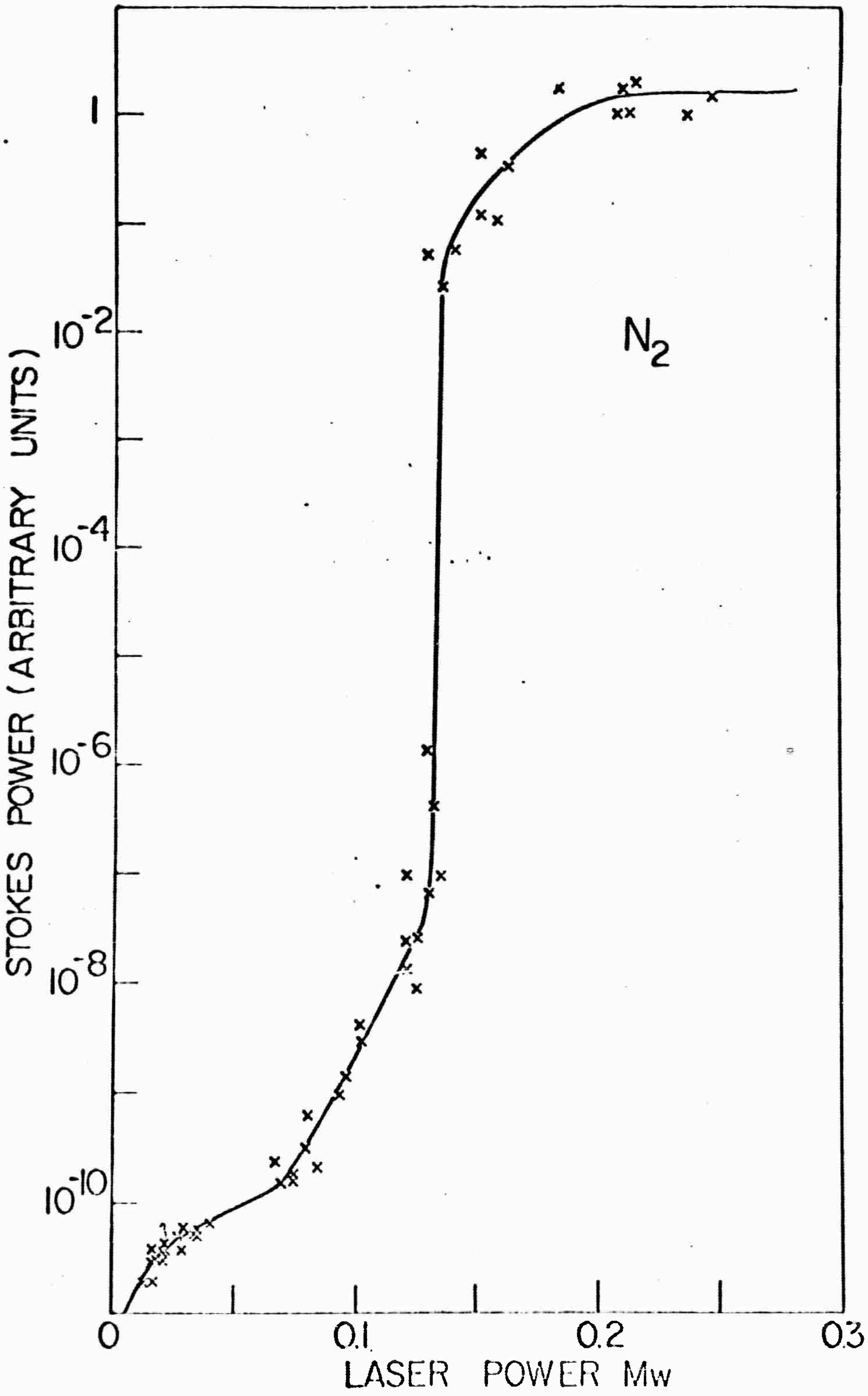
FIG. 3.

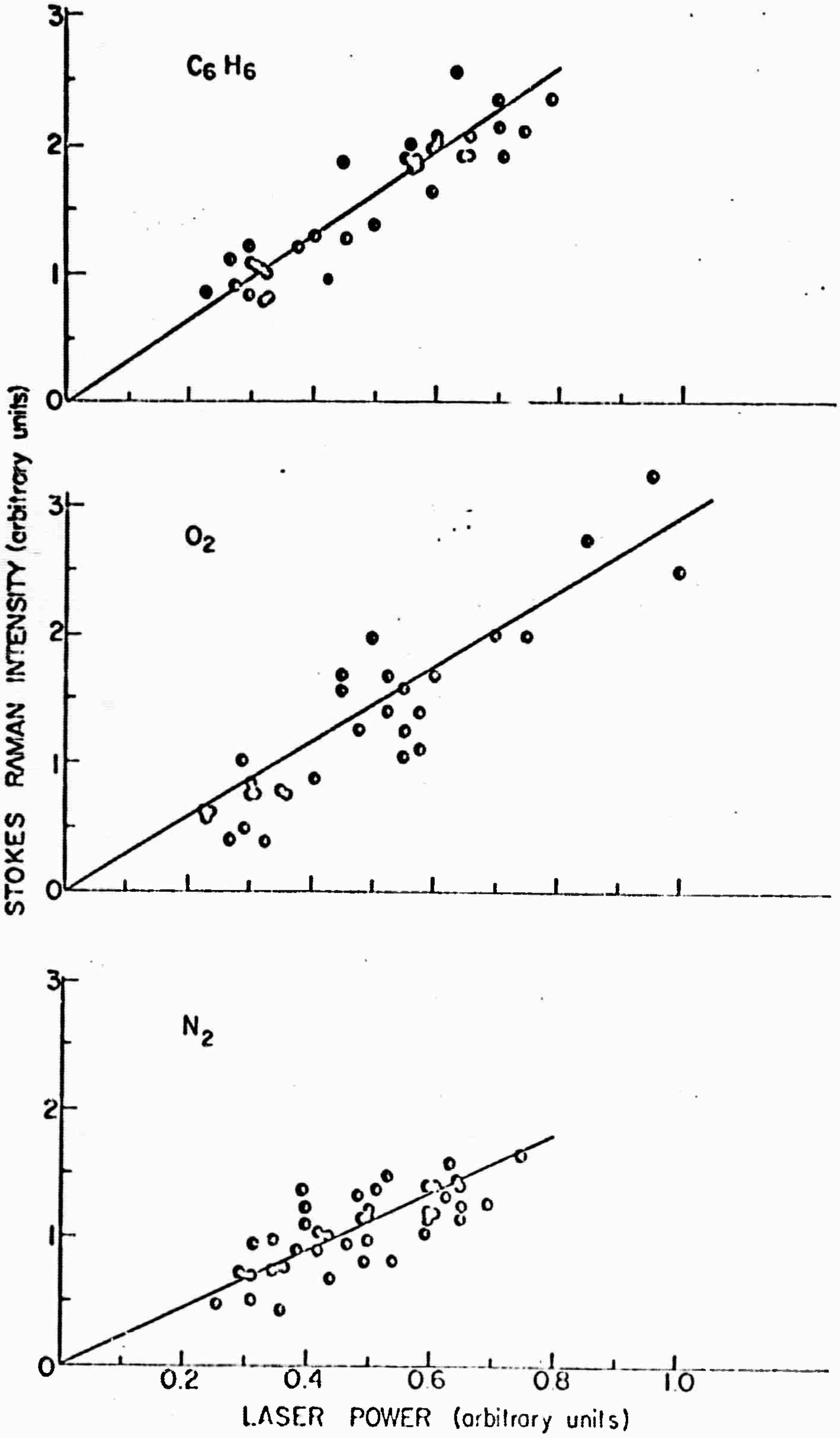
GRUN et al.


LIGHT TIGHT BOX
RUBY LASER
A.F.
P.D.
A.F.
D_1 (d = 0.6cm.)
L_1 (f = 50cm.)
A.F.
G.G.
P.D.
L_2
(f=5cm)
DEWAR
D_2
L_3
(f = 15cm.)
A.F.
F
SPEX
1700
G.G.
R.C.A.
7102
I STOKES
I LASER
TRIGGERING SIGNAL
TEKTRONIX 555

7. [illegible]


STOKES POWER (ARBITRARY UNITS)
10^-1
10^-3
10^-5
10^-7
10^-9
10^-11
O2
0
0.1
0.2
0.3
0.4
LASER POWER Mw


STOKES POWER (ARBITRARY UNITS)
1
10^{-2}
10^{-4}
10^{-6}
10^{-8}
10^{-10}
N_2
0
0.1
0.2
0.3
LASER POWER Mw


C_6H_6
O_2
N_2
STOKES RAMAN INTENSITY (arbitrary units)
LASER POWER (arbitrary units)
0
1
2
3
0.2
0.4
0.6
0.8
1.0

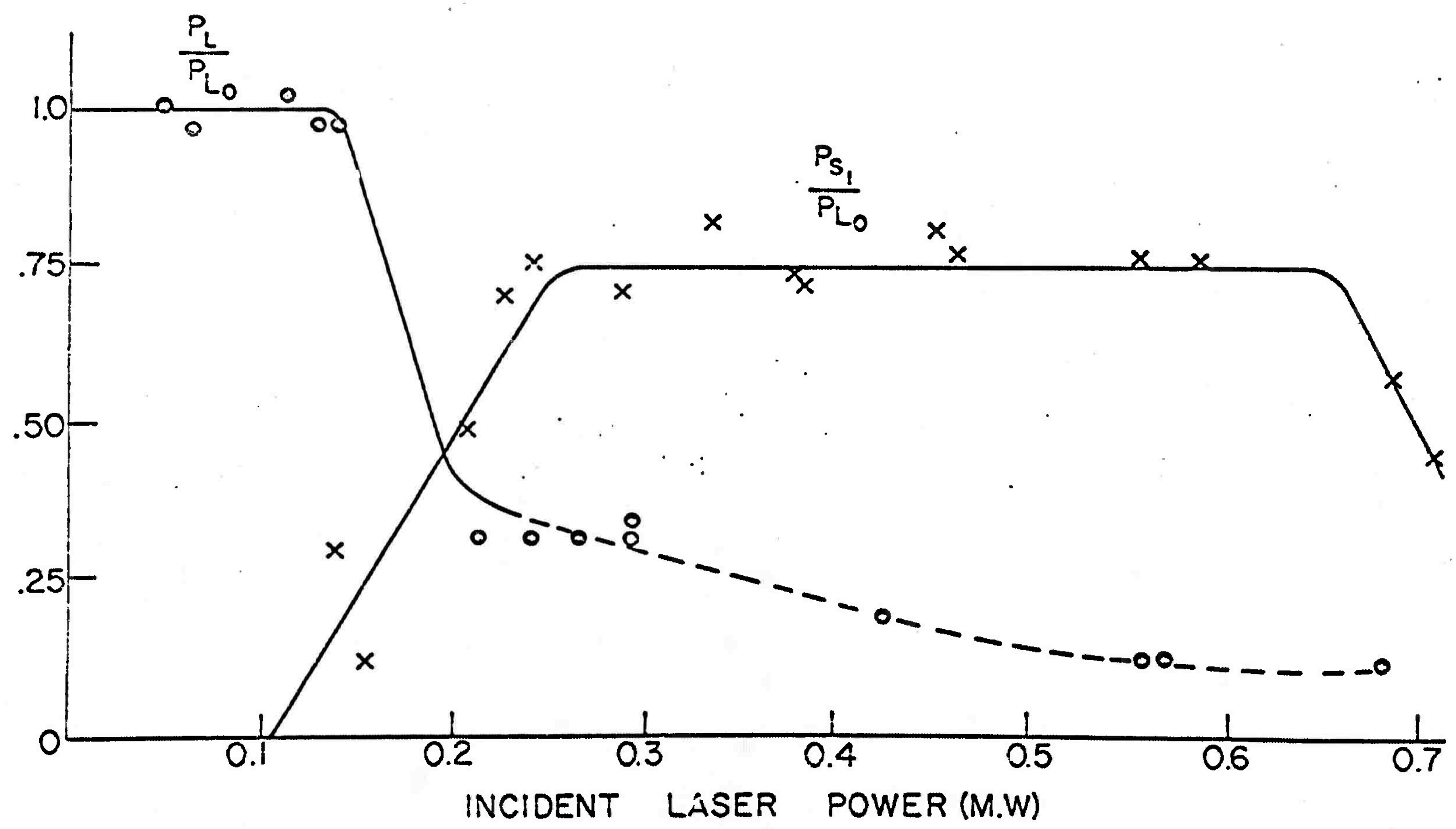


FIG 8 GRUN et al

DOCUMENT CONTROL DATA - R&D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY *(Corporate author)*

Department of Physics,
University of Toronto,
TORONTO 5, Ontario, Canada

2a. REPORT SECURITY CLASSIFICATION

UNCLASSIFIED

2b. GROUP

3. REPORT TITLE

Intensity and Gain Measurements of the Stimulated Raman Emission in Liquid O_2 and N_2

4. DESCRIPTIVE NOTES *(Type of report and inclusive dates)*

Semi-Annual Report No. 7 1 June 1968 - 1 January 1969

5. AUTHOR(S) *(Last name, first name, initial)*

Stoicheff, Boris P. Grun, J. Bernard
McQuillan, Archibald K,

6. REPORT DATE	**7a. TOTAL NO. OF PAGES**	**7b. NO. OF REFS**
December 1968	31	30

8a. CONTRACT OR GRANT NO.

Nonr-5012(00) M2

b. PROJECT NO.

NR 015-813/4-14-65

c.

Authorization ARPA Order No. 306

d.

9a. ORIGINATOR'S REPORT NUMBER(S)

Semi Annual Report No. 7

9b. OTHER REPORT NO(S) *(Any other numbers that may be assigned this report)*

10. AVAILABILITY/LIMITATION NOTICES

11. SUPPLEMENTARY NOTES

12. SPONSORING MILITARY ACTIVITY

Office of Naval Research and
Advanced Research Projects Agency

13. ABSTRACT

In liquid O_2 and N_2 the threshold for stimulated Raman emission is found to be much lower than for other nonlinear processes. Thus it is possible to make reliable measurements of the intensity of Raman emission over a large range of incident laser power by using a simple longitudinal geometry. Several distinct regions of emission were investigated, including normal Raman scattering, exponential gain, onset of oscillation and saturation. There is good agreement with theory.

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Raman Intensity and Gain						
Stimulated Raman Emission in O_2 and N_2						
Raman conversion efficiency in liquid N_2						

INSTRUCTIONS

1. ORIGINATING ACTIVITY: Enter the name and address of the contractor, subcontractor, grantee, Department of Defense activity or other organization *(corporate author)* issuing the report.

2a. REPORT SECURITY CLASSIFICATION: Enter the overall security classification of the report. Indicate whether "Restricted Data" is included. Marking is to be in accordance with appropriate security regulations.

2b. GROUP: Automatic downgrading is specified in DoD Directive 5200.10 and Armed Forces Industrial Manual. Enter the group number. Also, when applicable, show that optional markings have been used for Group 3 and Group 4 as authorized.

3. REPORT TITLE: Enter the complete report title in all capital letters. Titles in all cases should be unclassified. If a meaningful title cannot be selected without classification, show title classification in all capitals in parenthesis immediately following the title.

4. DESCRIPTIVE NOTES: If appropriate, enter the type of report, e.g., interim, progress, summary, annual, or final. Give the inclusive dates when a specific reporting period is covered.

5. AUTHOR(S): Enter the name(s) of author(s) as shown on or in the report. Enter last name, first name, middle initial. If military, show rank and branch of service. The name of the principal author is an absolute minimum requirement.

6. REPORT DATE: Enter the date of the report as day, month, year; or month, year. If more than one date appears on the report, use date of publication.

7a. TOTAL NUMBER OF PAGES: The total page count should follow normal pagination procedures, i.e., enter the number of pages containing information.

7b. NUMBER OF REFERENCES: Enter the total number of references cited in the report.

8a. CONTRACT OR GRANT NUMBER: If appropriate, enter the applicable number of the contract or grant under which the report was written.

8b, 8c, & 8d. PROJECT NUMBER: Enter the appropriate military department identification, such as project number, subproject number, system numbers, task number, etc.

9a. ORIGINATOR'S REPORT NUMBER(S): Enter the official report number by which the document will be identified and controlled by the originating activity. This number must be unique to this report.

9b. OTHER REPORT NUMBER(S): If the report has been assigned any other report numbers *(either by the originator or by the sponsor)*, also enter this number(s).

10. AVAILABILITY/LIMITATION NOTICES: Enter any limitations on further dissemination of the report, other than those imposed by security classification, using standard statements such as:

(1) "Qualified requesters may obtain copies of this report from DDC."

(2) "Foreign announcement and dissemination of this report by DDC is not authorized."

(3) "U. S. Government agencies may obtain copies of this report directly from DDC. Other qualified DDC users shall request through
________________."

(4) "U. S. military agencies may obtain copies of this report directly from DDC. Other qualified users shall request through
________________."

(5) "All distribution of this report is controlled. Qualified DDC users shall request through
________________."

If the report has been furnished to the Office of Technical Services, Department of Commerce, for sale to the public, indicate this fact and enter the price, if known.

11. SUPPLEMENTARY NOTES: Use for additional explanatory notes.

12. SPONSORING MILITARY ACTIVITY: Enter the name of the departmental project office or laboratory sponsoring *(paying for)* the research and development. Include address.

13. ABSTRACT: Enter an abstract giving a brief and factual summary of the document indicative of the report, even though it may also appear elsewhere in the body of the technical report. If additional space is required, a continuation sheet shall be attached.

It is highly desirable that the abstract of classified reports be unclassified. Each paragraph of the abstract shall end with an indication of the military security classification of the information in the paragraph, represented as *(TS)*, *(S)*, *(C)*, or *(U)*.

There is no limitation on the length of the abstract. However, the suggested length is from 150 to 225 words.

14. KEY WORDS: Key words are technically meaningful terms or short phrases that characterize a report and may be used as index entries for cataloging the report. Key words must be selected so that no security classification is required. Identifiers, such as equipment model designation, trade name, military project code name, geographic location, may be used as key words but will be followed by an indication of technical context. The assignment of links, rules, and weights is optional.