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FINAL TECHNICAL REPORT

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STUDY OF LOSS MECHANISMS AND STIMULATED RAMAN EMISSION

1 May 1965 through 3 December 1965

by

C. C. Wang

Contract No. Nonr-4850(00)

ARPA Order 306 Program Code No. 4730

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A SUBSIDIARY OF Ford Motor Company
APPLIED RESEARCH LABORATORY
BLUE BELL, PENNSYLVANIA

STUDY OF LOSS MECHANISMS AND STIMULATED RAMAN EMISSION

C. C. Wang

Philco Applied Research Laboratory
Blue Bell, Pa.

Final Technical Report

1 May 1965 to 31 December 1965

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March 1966

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ABSTRACT

This report summarizes work done at Philco Applied Research Laboratory under Contract Nonr-4850(00). Extensive measurements were taken on the conversion efficiency and threshold of stimulated Raman emission in benzene, nitrobenzene, toluene, and carbon disulfide. It has been shown that the self-focusing action of a laser beam is responsible for the onset of stimulated Raman emission.

1. INTRODUCTION

This report summarizes work done at Philco Applied Research Laboratory under Contract Nonr-4850(00). The contract work was begun on May 1, 1965; it was terminated on December 31, 1965 upon mutual agreement between Philco and the contracting officer. As of January 1, 1966, the personnel originally assigned to the program were transferred from Philco Applied Research Laboratory to the Scientific Laboratory of Ford Motor Company.

During the period of the contract, experimental and theoretical considerations were given to the single-mode operation of a Q-switched ruby laser. This part of the work is presented in Section 5 in the form of a paper submitted for publication in Journal of Applied Physics. To fulfill the objectives of the contract, extensive measurements were taken on the conversion efficiency and threshold of stimulated Raman emission in benzene. This is summarized in Section 4, in the form of a paper to be published in the April 1966 issue of the Journal of Applied Physics.

The contract work at Philco has led to additional work on the same subject performed with Ford funding. Some of this work is included here in Section 2 in the form of a paper published in Physical Review Letters, and in Section 3 in the form of a paper submitted for publication in Applied Physics Letters. It must be emphasized that the materials in Sections 2 and 3 were <u>not</u> obtained under contract. They are included in this report because of their direct bearings on the objectives of the contract, and consequently are included for information only.

The enclosed publications fully summarize the progress made in the research done under the contract. We believe these publications represent significant contributions to the field.

2. LENGTH-DEPENDENT THRESHOLD FOR STIMULATED RAMAN EFFECT AND SELF-FOCUSING OF LASER BEAMS IN LIQUIDS.*

(Published in Phys. Rev. Letters 16, 344 (1966)

* The work reported in this section was not done under contract. It is included here for information only.

LENGTH-DEPENDENT THRESHOLD FOR STIMULATED RAMAN EFFECT AND SELF-FOCUSING OF LASER BEAMS IN LIQUIDS

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Recent experiments[1-3] have revealed the formation of high-intensity filaments in the laser beam during its passage through Raman-active liquids. Such high-intensity filaments are believed to be formed by the self-focusing action of laser beams[3,4], which in turn is due to the intensity dependent index of refraction[5,6]. It has been noted[3] that the experimental threshold for stimulated Raman emission appears to be determined not so much by the value of the Raman susceptibility, but rather by the self-focusing capability of the liquids. In particular, the onset of stimulated Raman emission has been observed to occur only after the beam has traveled some distance through the liquid[1,3], and it has been proposed[4] that this is the distance required for self-focusing to develop. In this letter we report our length-dependent threshold data for stimulated Raman emission in several liquids, and show that these data can be interpreted in terms of the predicted dependence of self-focusing. By extrapolating the results to liquid cells of infinite length, we obtain values of critical power for self-trapping[7] of the laser beam in liquids. We believe this is the first measurement from which the critical power for self-trapping has been deduced.

The self-focusing effect can be understood by considering the diffraction of a laser beam in material which exhibits intensity dependent index of refraction. For normal dielectrics, the index increases with increasing intensity so that the phase velocity decreases with increasing intensity. A lens effect is thus produced whereby the rays move toward the

region of highest intensity and the intensity there increases. This increase in intensity is accompanied by a reduction in effective beam width, and continues until it is limited by other factors. A threshold exists for the onset of self-focusing as it must overcome the spreading of the beam due to diffraction. Chiao, Garmire and Townes[7] have predicted that a light beam may be trapped at any arbitrary diameter and will thus not spread. They have further predicted that self-trapping occurs at a critical power level independently of the beam diameter. Experiments[3,8] with high power laser beams have shown that high intensity filaments of 20 to 80 in diameter are formed, presumably due to self-focusing. While it is not yet understood which factors determine the size of the filaments, the distance required for the establishment of these filaments should depend very little on the terminal filament size if the beam is reduced in diameter by an appreciable factor; this is particularly so because the nonlinear nature of the focusing causes the beam diameter to decrease more rapidly the more the beam diameter decreases. This distance has been calculated by Kelley,[4] and has been referred to as the self-focusing length ℓ,

$$\ell = \frac{n_o}{4} \left(\frac{a^2}{f}\right) \left(\frac{c}{n_2}\right)^{\frac{1}{2}} \Big/ \left[\sqrt{P} - \sqrt{P_{cr}}\right] \tag{1}$$

Here n_o is the linear index of refraction, c is the speed of light in vacuum, n_2 is related to the dc change in index as defined in Reference 7, P is the input laser power, and P_{cr} is the critical power for cylindrical beam trapping,[7]

$$P_{cr} = \frac{(1.22\lambda)^2 \, c}{256 \, n_2} \tag{2}$$

Equation (1) has been modified slightly from that used by Kelley.[4] Here f is the ratio of the radius of the beam, a, to a characteristic transverse

radius of curvature of the laser intensity; it is introduced here as a parameter to allow for deviations from an equiphase Gaussian intensity profile. f = 1 for such a Gaussian beam assumed by Kelley[4].

Equation (1) may be rewritten as

$$\sqrt{P} = \sqrt{P_{cr}} + \alpha/\ell \tag{3}$$

where

$$\alpha = \frac{n_o}{4}\left(\frac{a^2}{f}\right)\left(\frac{c}{n_2}\right)^{\frac{1}{2}} \tag{4}$$

Equation (3) gives the threshold power for the formation of filaments in a liquid column of a given length. Thus if P is to be taken as the observed threshold for stimulated Raman emission, the plot of $\sqrt{P}$ vs. $1/\ell$ for various materials should yield straight lines. The intercepts on the ordinate of these straight lines should give the values for P_{cr}, whereas the slopes of these lines should be proportional to (a^2/f) and could thus be used to determine the ratio f of radii introduced in Equation (1).

The experimental setup used to obtain the threshold data for stimulated Raman emission has been described elsewhere[9]. The observed Stokes power as a function of the incident laser power was plotted over ten orders of magnitude in a log-log plot. The value of the threshold laser power was then determined by observing the sharp break in the curve as transition took place from the linear spontaneous region to the stimulated region. The relative accuracy of the threshold values thus obtained is typically $\pm$ 20% for most liquid cells, and slightly higher for very short cells. The absolute power measurement is estimated to be accurate to $\pm$ 50%. Figure 1 shows these threshold data for benzene, toluene, and nitrobenzene. One observes that Equation (1) is well satisfied for all three liquids investigated.

With the data compiled in Reference 5 for n_2, the values of P_{cr} calculated from Equation (2) and from Equation (2) of Reference 7 are listed in Table I along with the observed values. Note that the power calculated from our Equation (2) is one-fourth as much as the critical power deduced in Reference 7 on the basis of elementary considerations. The observed values fall between these two values.

Also included in Table I are the values of (f/a^2) obtained both in benzene and in nitrobenzene. These two values for (f/a^2) are found to be the same within experimental accuracy. This is to be expected since (f/a^2) is characteristic of the laser beam.

We have studied experimentally the intensity profile of the incident laser beam. The laser beam was found to contain four nearly circular spots approximately equal in intensity and each about (0.1 ± 0.02) cm in diameter. Experiments with two of the spots blocked gave the same results within experimental error. The intensity distribution of these spots was asymmetrical and exhibited curvature smaller than the curvature for a Gaussian beam. Assuming a circular spot, one obtains from Table I values of f near two. Previously[3], f has been estimated to be ≈ 10. Beams whose curvature is greater than the curvature for a Gaussian beam will have larger values of f and thus have shorter self-focusing length. Multimode effects should also increase the f - values. On the other hand, possible two photon absorption tends to impede the self-focusing action, and will reduce the f - values.

We have also performed threshold measurements with laser beams of three different cross sections. The result for benzene is plotted in Figure 2. Here the experimental points are fitted to three straight lines with common intercept on the vertical axis; the slope of these lines are

seen to increase in linear proportion to the cross sectional area of the beam. This is in agreement with the predictions that the critical power for self-trapping is independent of the beam cross section, and that the quantity $(\sqrt{P} - \sqrt{P_{cr}})$ expressing the threshold for Raman laser action should increase as the square of the radius of the beam. Qualitative results of a similar nature have also been observed by McClung[11].

The author is indebted to R. W. Terhune for several very useful discussions, and to G. W. Racette for assistance in conducting the experiments.

TABLE I

Comparison of critical powers for self-focusing and of ratios f of radii.

Material	Critical Power P_{cr} (MW)			(f/a^2) (cm^2)
	Observed	Calculated [a]	Calculated [b]	
Benzene	0.064	0.021	0.085	7.9×10^2
Nitrobenzene	0.019	0.005	0.021	8.3×10^2
Toluene	0.055	—	—	—

a. Reference 4

b. Reference 7

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FIGURE CAPTIONS

Figure 1. Plot of the square root of the threshold laser power for stimulated Raman emission as a function of the inverse of the cell length for benzene, toluene, and nitrobenzene.

Figure 2. Same plot as Figure 1 for benzene, but with beams of different cross sections. The beam cross section used is (a) 1.25×10^{-2} cm^2, (b) 0.78×10^{-2} cm^2, and (c) 0.48×10^{-2} cm^2. This corresponds to a ratio of 1.6: 1: 0.615 among the cross sections, as may be compared with the ratio of 1.5: 1: 0.6 among the observed slopes.

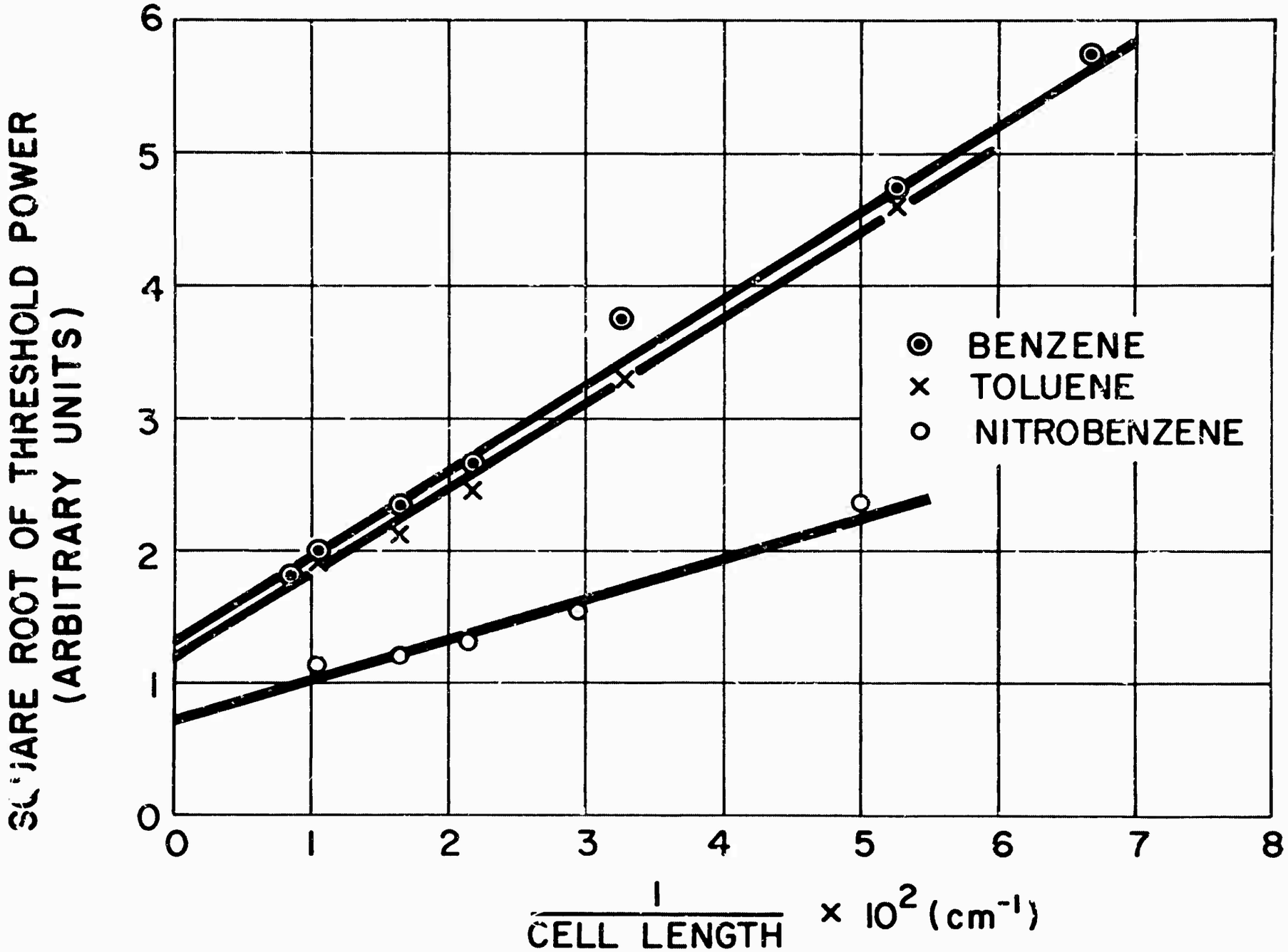
SQUARE ROOT OF THRESHOLD POWER
(ARBITRARY UNITS)
0
1
2
3
4
5
6
0
1
2
3
4
5
6
7
8
BENZENE
TOLUENE
NITROBENZENE
1
CELL LENGTH
× 10² (cm⁻¹)

Figure 1

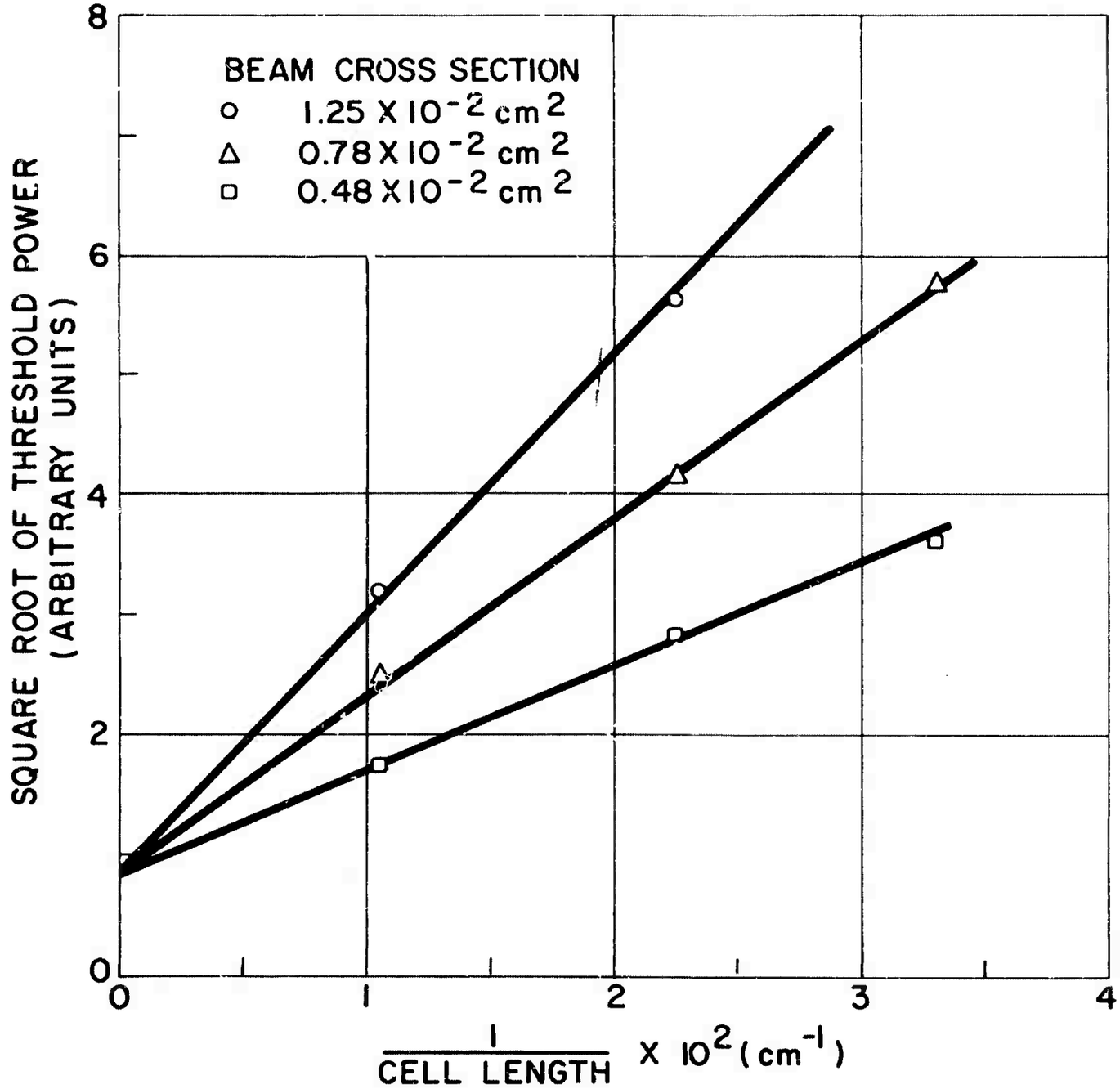
BEAM CROSS SECTION
○ 1.25 X 10⁻² cm²
△ 0.78 X 10⁻² cm²
□ 0.48 X 10⁻² cm²
SQUARE ROOT OF THRESHOLD POWER
(ARBITRARY UNITS)
0
2
4
6
8
0
1
2
3
4
1 / CELL LENGTH X 10² (cm⁻¹)

Figure 2

3. EFFECT OF LINEAR ABSORPTION ON SELF-FOCUSING OF LASER BEAM IN CS_2*

(Submitted for publication in Applied Physics Letters)

* The work reported in this section was not done under contract. It is included here for information only.

EFFECT OF LINEAR ABSORPTION ON SELF-FOCUSING OF LASER BEAM IN CS_2

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The self-focusing of a laser beam in liquids has been the subject of several recent publications[1-8]. It has been shown[7] that the length-dependent threshold for stimulated Raman emission in benzene, nitrobenzene and toluene can be interpreted in terms of the predicted dependence of self-focusing. This letter reports the effect of linear absorption on the threshold for self-focusing in CS_2. The linear absorption coefficient α in CS_2 was varied from $0.002 cm^{-1}$ to $0.125 cm^{-1}$ by dissolving in CS_2 a controlled amount of iodine. The experimental results have been found to be in fair agreement with the theoretical prediction. It is of particular interest to note that with a peak power of 1 MW from our unfocused laser beam, no effect attributable to two-photon absorption was observed, although CS_2 exhibits the strongest two photon absorption[9] among all the known Raman active liquids.

The effect of linear absorption on the self-focusing of a laser beam can be predicted by including a linear loss term in the nonlinear wave equation. Using the approximations made by Kelley[5], we obtain the following expression for the threshold power, P, for self-focusing to occur in a cell of length ℓ in the presence of linear absorption:

$$(P^{\frac{1}{2}} - P_{cr}^{\frac{1}{2}}) / \alpha A = \sqrt{2} \Big/ \left[1 - (1+2\alpha\ell)e^{-2\alpha\ell}\right]^{\frac{1}{2}} \qquad (1)$$

Here P_{cr} is the critical power for cylindrical beam trapping[10], and $A = (n_o/4)\ (a^2/f)\ (c/n_2)^{1/2}$. n_o is the linear index of refraction, c is the speed of light in vacuum, n_2 is related to the dc change in index as defined in Ref. 1, and f is the ratio of the radius a of the beam to a

characteristic transverse radius of the laser intensity. When the linear absorption is small such that $\alpha\ell \ll 1$, Eq. (1) reduces to the form appropriate for a lossless medium,

$$P^{1/2} - P_{cr}^{1/2} = A/\ell \qquad (2)$$

which describes a linear relation between $P^{1/2}$ and $1/\ell$. On the other hand, when the absorption is large and $\alpha\ell \gg 1$, Eq. (1) is seen to approach asymptotically a constant value

$$P^{1/2} - P_{cr}^{1/2} = \sqrt{2}\,\alpha A \qquad (3)$$

Equation (3) states that in a long column of lossy medium, the threshold power for self-focusing is independent of the cell length and is equal to that corresponding to a cell $1/\sqrt{2}\alpha$ in length. This is to be expected since the effective self-focusing length must be short compared to the absorption length for self-focusing to occur.

The experiments were done with the unfocused output from a Q-switched ruby laser. The laser beam provided a peak power of 2 MW and was found to contain two nearly circular spots approximately equal in intensity and each about (0.1 ± 0.02)cm in diameter. The threshold power for self-focusing was taken as the threshold for stimulated Raman emission[7].

Figure 1 shows the threshold data for CS_2 samples corresponding to different linear absorption. The experimental points obtained with the undoped CS_2 well satisfy Eq. (2) with a measured slope of $A = 3.2\sqrt{MW}$-cm and the vertical intercept $P_{cr}^{1/2} = 0.125\sqrt{MW}$. This gives a critical power of 15kW for self-trapping, as may be compared with the theoretical threshold of 9kW calculated from the known optical Kerr constant.[11]

For the iodine-doped, highly absorbing samples of CS_2, Eq. (1) was fitted to the experimental data with $P_{cr}^{1/2}$ taken as an undetermined parameter. This gave a value of 0.072, 0.105, and $0.125\sqrt{MW}$ for $P_{cr}^{1/2}$ corresponding to

α = 0.125, 0.076 and 0.036 cm^{-1} respectively. According to Eq. (1), the same value of $P_{cr}^{1/2}$ should have been obtained from the four curves. The difference in these values is significant, indicating that a discrepancy exists between Eq. (1) and the experiments. Eq. (1) was derived on the basis of several oversimplifying approximations particularly in regard to the effect of beam diffraction, and it would be desirable to perform computer calculations of the kind done by Kelley[5] to check on this point.

We have also considered the effect of two photon absorption on the self-focusing of a laser beam. Using the same approximations employed in deriving Eq. (1), it can be shown that in the presence of two photon absorption, Eq. (2) is modified to become

$$P^{1/2} - P_{cr}^{1/2} = A/(\ell - \beta) \tag{4}$$

where

$$\beta = (\tfrac{1}{2})k\, a^2\, (\epsilon_2'' / \epsilon_2'), \tag{5}$$

a is the radius of the beam, $k = 2\pi n_0/\lambda$, and $\epsilon_2 = \epsilon_2' + i\epsilon_2''$ is the complex coefficient of the intensity dependent dielectric constant. Thus the effect of two photon absorption is to increase the threshold for self-focusing, making it impossible in a distance shorter than a critical length β. This is understandable since in such short cells the threshold intensity required for self-focusing may already be so high that two photon absorption process dominates. For CS_2[9], with $(\epsilon_2''/\epsilon_2') \sim 10^{-2}$ and a 1mm diamter beam, one obtains $\beta \sim 1$cm. 4cm was the shortest cell length in which self focusing was observed. The small predicted deviation due to two photon absorption was not detected.

The authors are grateful to Dr. R. W. Terhune for many stimulating discussions during the course of the experiments.

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FIGURE CAPTION

Figure 1. Plot of the square root of the threshold laser power for self-focusing in CS_2 as a function of the inverse of the cell length. The straight line (d) is drawn through the experimental points obtained with the undoped CS_2. The solid curves are theoretical fits of Eq. (1) with A = 3.2 and the respective values of α, taking $P_{cr}^{1/2}$ as an undetermined parameter.

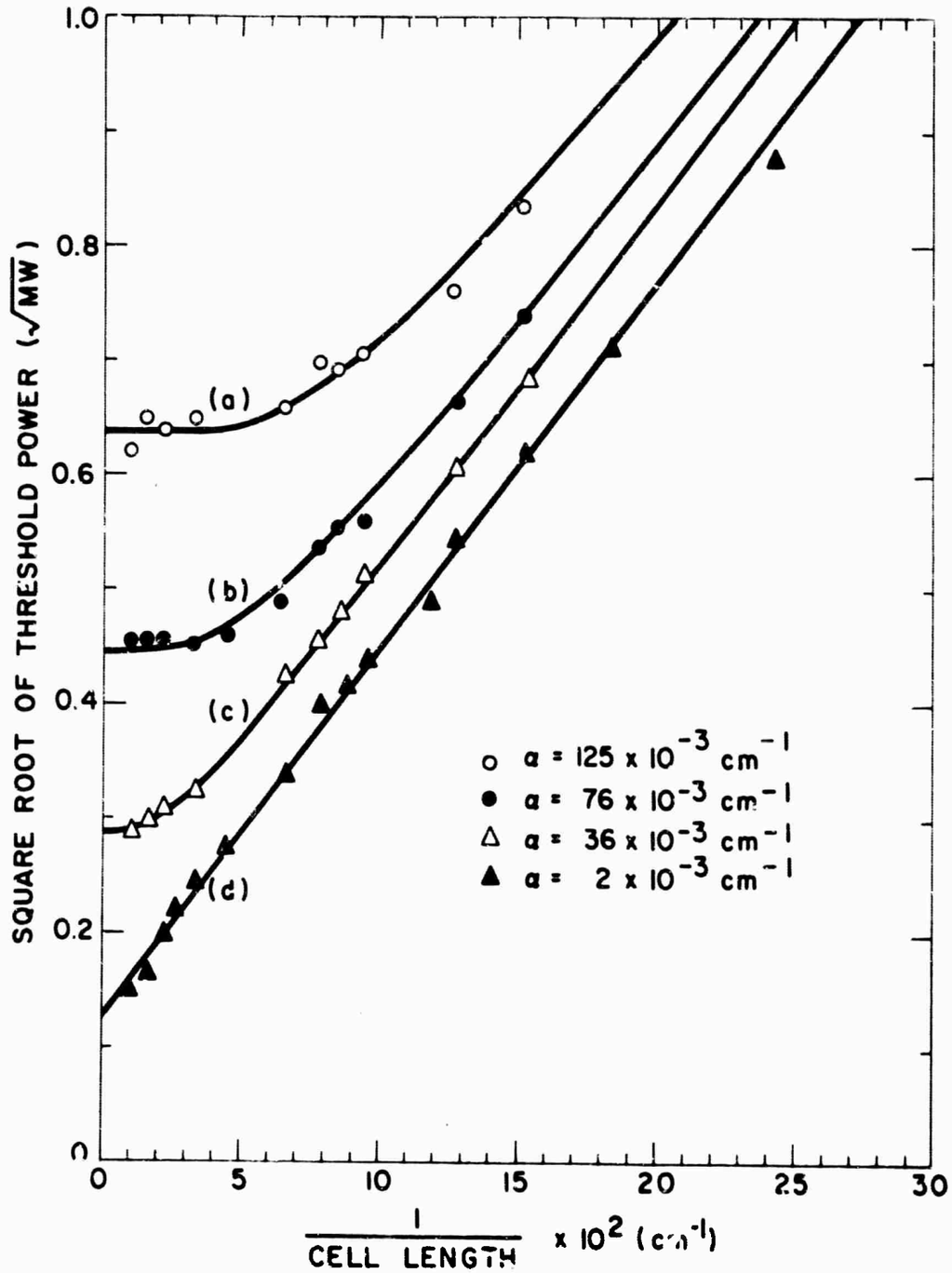
1.0
0.8
0.6
0.4
0.2
0
SQUARE ROOT OF THRESHOLD POWER ($\sqrt{MW}$)
(a)
(b)
(c)
(d)
○ α = 125 x 10^{-3} cm^{-1}
● α = 76 x 10^{-3} cm^{-1}
△ α = 36 x 10^{-3} cm^{-1}
▲ α = 2 x 10^{-3} cm^{-1}
0
5
10
15
20
25
30
$\frac{1}{\text{CELL LENGTH}}$ x 10^2 (cm^{-1})

Figure 1

4. LENGTH DEPENDENCE OF STIMULATED RAMAN EFFECT IN BENZENE

(To be published in Journal of Applied Physics)

LENGTH DEPENDENCE OF STIMULATED RAMAN EFFECT IN BENZENE*

By

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The existence of anomalously high gain in stimulated Raman scattering in liquids has been the subject of several recent publications[1-8]. According to the theory of Hellwarth,[9] the fractional conversion of the laser photons into the Stokes photons is given by

$$\frac{n_s}{n_{\ell o}} = \frac{1}{1 + (n_{\ell o}/n_{so})\, e^{gL}} \tag{1}$$

Here n_ℓ and n_s are the laser and Stokes photons per cm^2 per sec; the subscript "o" denotes the initial values; L is the length of the Raman cell, and g is the gain per unit length at the Stokes frequency. g is related to the peak differential scattering cross section σ in units of cm^2 per unit volume per steradian per unit wavelength through the equation

$$g = n_\ell \, \sigma \, \lambda_R^4 / c_R \tag{2}$$

where λ_R is the wavelength of light in the medium at the Stokes frequency, and c_R is the speed of light in the medium at this frequency. It has been observed both in nitrobenzene[1-6] and in benzene[3] that the experimental conversion efficiency curves do not fit Equation (1) with constant value of gain per unit length. Immediately above threshold, the gain per unit length for the Stokes

* This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Project Agency, the Office of Naval Research, and the Department of Defense.

intensity has been found to be one or two orders of magnitude higher than the theoretical value predicted by Equation (2) using the cross section obtained in normal (spontaneous) Raman scattering experiments. As the laser intensity is increased further beyond the threshold, the gain began to decrease and approached the theoretical value as large fractional power conversion is reached. Although several possible mechanisms have been suggested to explain the above-mentioned discrepancy, it is clear that more theoretical and experimental work is required to find the correct explanation.

The existence of a threshold, both in the exciting laser intensity and in the length of the liquid cell, was first noted by Eckhardt et al,[10] and subsequently by Stoicheff.[11] Recently, Lallemand and Bloembergen[8] measured the Stokes intensity as a function of cell length at constant laser intensity, and a sharp break indicating the onset of stimulated emission was noted. Bret and Mayer[3] observed that the laser intensity at threshold was a decreasing function of cell length although such functional dependence was not explicitly determined. On the other hand, there have been some discrepancies among the results on conversion efficiencies reported to date. This is perhaps because of the differences in the laser beams used in the various experiments so that results obtained under dissimilar conditions were compared. It was thus felt desirable to conduct a comprehensive study of the experimental aspects of the stimulated Raman effect under specified conditions. We wish to report in this note our quantitative threshold and conversion efficiency measurements for stimulated Raman emission in benzene, with cell length ranging from 4 cm to 150 cm. We believe that we have established for the first time the threshold laser intensity for stimulated emission as a function of the cell length.

In the region of high power conversion, we have observed that the incident laser beam after passage through the Raman cell was definitely distorted. We have also found that the anomalously high gain in the stimulated region is dependent upon the cell length.

To perform such measurements, unfocused output from a giant pulse laser was used. The laser was Q-switched by a rotating prism, providing a 60 mJ pulse with 30 nsec half-width. Inclusion of a sapphire mode selector[12-14] in the cavity ensured that the output contained only one longitudinal mode with its spectral width less than 0.02 cm^{-1}. Occasionally two longitudinal modes separated by 0.45 cm^{-1} were observed. The output consisted of two parallel beams each about 2 mm in diameter and was collimated to better than 1 mrad. The quality of the beam was substantiated by the fact that 15% of the beam intensity was sufficient to create dielectric breakdown in air when brought to focus by an one-inch focal length lens. The liquid cells were carefully tilted so that their windows were a few degrees off from being perpendicular to the laser beam. The detection of the Stokes power over about ten orders of magnitude was facilitated through use of narrow-band interference filter and suitable combination of Corning glass filters.

Below threshold, the spontaneous Stokes power in the forward direction was measured as a function of the incident laser intensity. The photomultiplier accepted the Stokes beam over a solid angle of 2.5×10^{-3} steradian. Figure 1 shows the linear relationship between the Stokes and the laser intensities in the spontaneous region, from which the differential Raman scattering cross section at 6943 Å is calculated to be 5.08 cm^2 per unit volume per steradian per unit wavelength. Assuming a λ^4 dependence, this corresponds to an absolute

cross section of 0.76×10^{-28} cm^2 at 6328Å. Using a cw He-Ne laser operating at 6328Å, Damen et al[15] obtained a value of 0.56×10^{-28} cm^2 for benzene. Our cross section is probably good to within ± 50%, so that these two values agree within experimental accuracy. Comparable results have also been obtained by Bret[16] in the forward direction, and by McClung and Weiner[17] in a direction making an angle of 90° with the laser beam.

Figure 2 shows the threshold intensity for stimulated Raman scattering in benzene as a function of the cell length. For large L, i.e., $L \gtrsim 30$ cm, the threshold intensity is seen to increase as L^{-1}, whereas for small L, the threshold intensity increased much faster than L^{-1}. Accurate determination of threshold intensity in short cells was made difficult because of the strong filter fluorescence at higher laser intensities. In long cells, the effect of beam divergence was not negligible; in obtaining Figure 2, this effect was taken into account in a straightforward manner.

In the stimulated region, the Stokes beam emerged from the Raman cell in a cone of approximately 10^{-5} steradian. Spectroscopic analysis using a B & L 2m grating spectrograph indicated the existence of the first Stokes and the second Stokes radiation, the latter being usually one or two orders down in intensity from the former. With our maximum laser intensity, no third Stokes radiation of sufficient intensity to produce a developable image in the spectral plate was observed. The half-width of the Stokes line was measured with a Fabry-Perot interferometer, and was found to be about 0.2 cm^{-1}. In the region of high power conversion, the laser beam after passage through the Raman cell was noticeably distorted; such distortion was evident from photographs taken at

a point beyond the exit end of the cell. The maximum conversion efficiency into the first Stokes was measured with a calibrated ballistic thermopile to be in excess of 45%. This value of conversion efficiency was confirmed by measuring the depletion of laser intensity after passage through the cell. By comparing the results with and without the Raman cell inserted in the path of the laser beam, a depletion of about 50% in laser intensity was noted.

Figure 3 shows the Stokes intensity generated in various cells as a function of the laser intensity.[18] Using the value of 5.08 cm^2 derived from the data presented in Figure 2 for the cross section, one calculated from Equation (2) a theoretical gain of 5×10^{-3} cm^{-1} for a laser power of 1 MW/cm^2. The average gain in the region not too far above the threshold may be estimated from the data presented in Figure 3 and the measured spontaneous Stokes intensity at threshold. The average gain computed on the basis of one-pass amplification[1] for the 90 cm, 45 cm, 15 cm, and 10 cm cells are respectively 51, 48, 17, and 10 times higher than the corresponding theoretical gain. This average gain in the stimulated region is thus seen to increase monotonically with the cell length, and correlates closely with the threshold curve of Figure 2. We are currently engaged in similar experimental studies on other Raman liquids and the result will be the subject for subsequent publication.

It is a pleasure to acknowledge the constant assistance of G. W. Racette and G. T. Mitchell throughout the experiments.

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18. The linear absorption in benzene was measured to be about 10^{-3} cm^{-1}. For the cell lengths investigated, this amount of absorption is too small to bring about any cross-over of curves corresponding to cells of different length. The author is indebted to Prof. C. H. Townes for pointing out a numerical error in the preliminary plot of these conversion efficiency data.

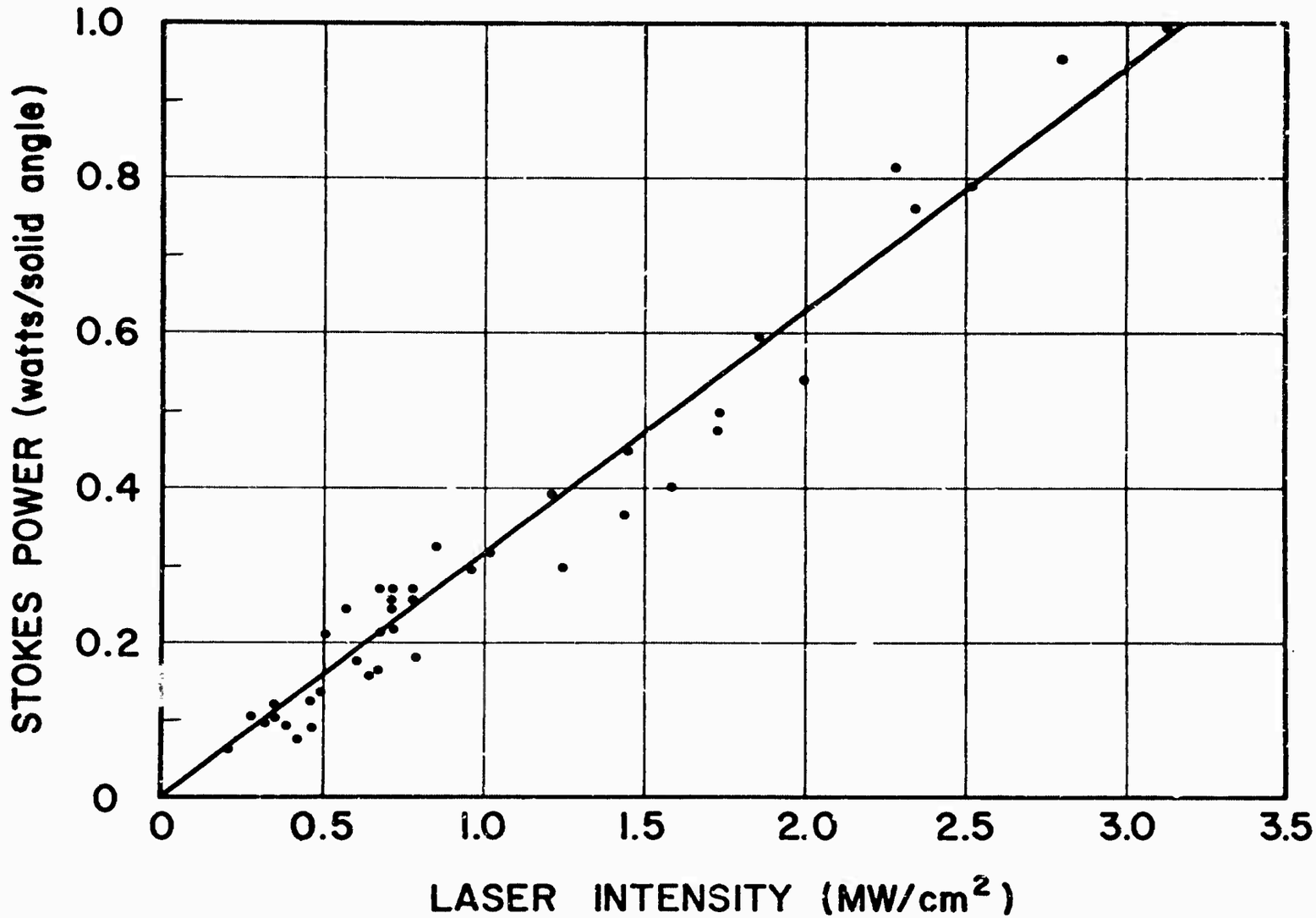

Figure 1

Spontaneous Stokes power from a 45 cm benzene cell. The measurements were made over a solid angle of 2.5×10^{-3} steradian, and the laser beam cross section was 0.08 cm^2.

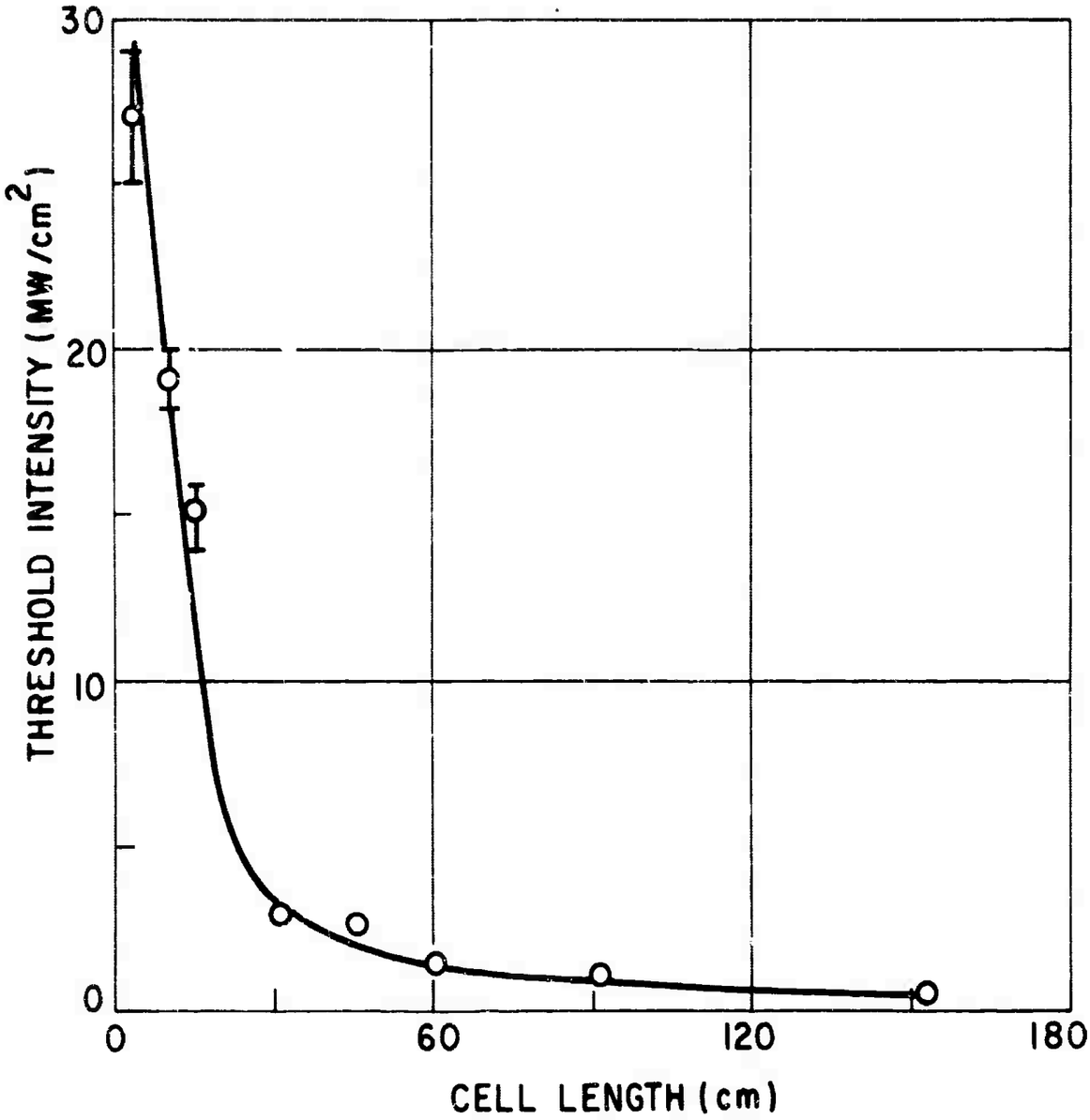

Figure 2

Threshold intensities for stimulated Raman scattering in benzene.

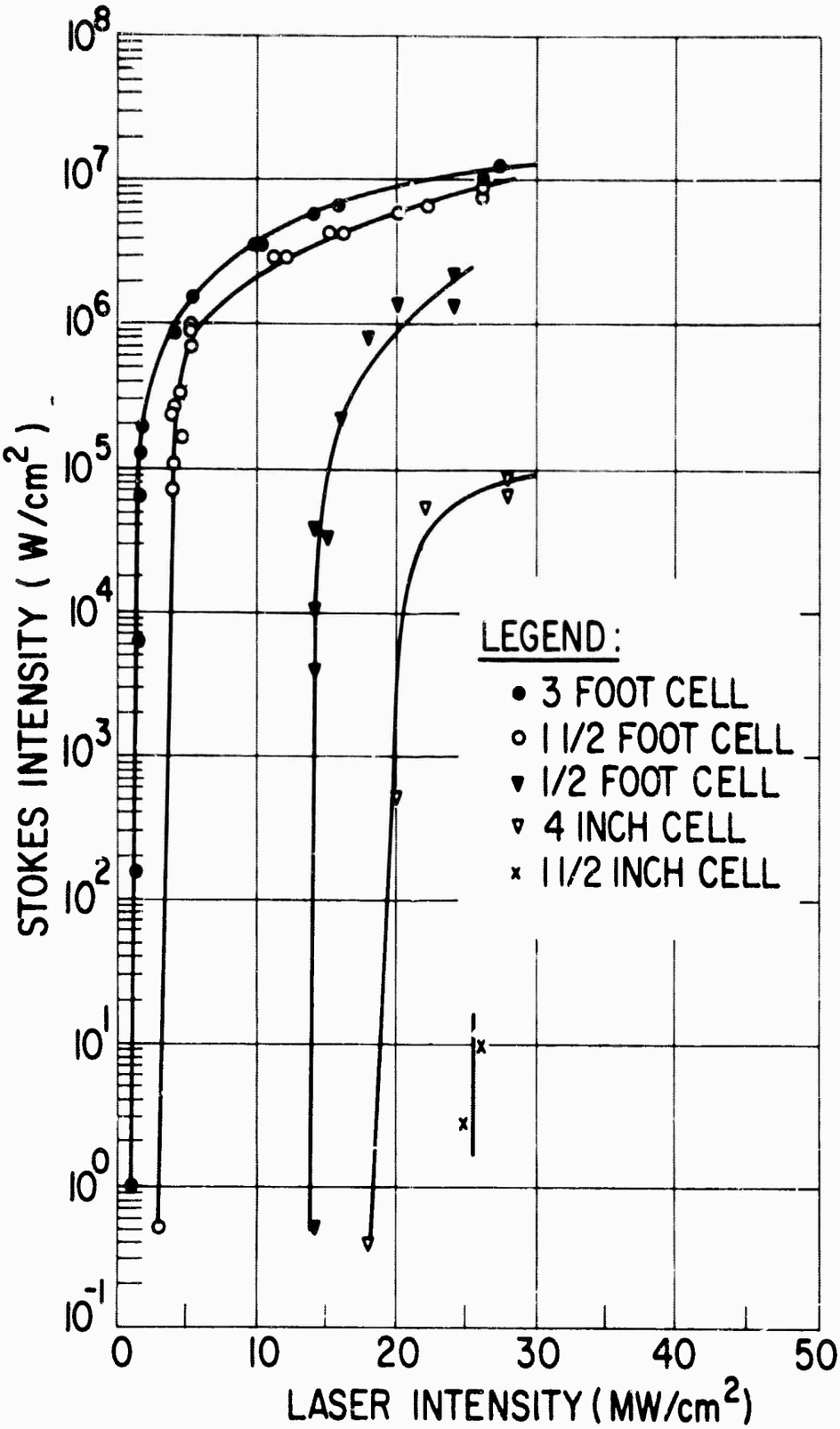

Figure 3

Plots of Stokes intensity vs. laser intensity for benzene cells of different length.

5. MODE SELECTION AND LINEWIDTH NARROWING IN GIANT PULSE LASERS

(Submitted for publication in Journal of Applied Physics)

MODE SELECTION AND LINEWIDTH NARROWING IN GIANT PULSE LASERS*

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ABSTRACT

The rate equations governing the multimode giant pulse laser are examined in regard to the total emitted energy in the pulse and the linewidth narrowing. It is found that the total energy from a multimode pulse is approximately the same as that from a single-mode pulse, and that the linewidth narrowing is determined by the number of back and forth reflections of the amplified spontaneous radiation within the cavity, and by the gain (or loss) profile of the laser. Experimental results with a transmission mode selector are discussed.

* This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Project Agency, the Office of Naval Research, and the Department of Defense.

INTRODUCTION

The attainment of optical giant pulses by switching of the cavity regeneration was proposed by Hellwarth[1], and subsequently realized by McClung and Hellwarth[2]. The theory and operation of the giant pulse laser have been discussed in the literature[3-9]. However, the theory so far developed is strictly applicable only for the single-mode operation and is admittedly inadequate to describe the behavior of a practical giant pulse laser, which usually is a multimode device because of its characteristic high initial excitation. While the single-mode operation is obtainable through incorporation of suitable mode selection techniques[10,11], the spectral characteristics of a giant pulse ruby laser without mode selection are far from being ideal. The techniques of mode selection are well described[12-15], but an analytical account pertaining to mode selection and linewidth narrowing in a multimode giant pulse laser has been lacking. It is the purpose of this paper to attempt such an analysis.

The occurrence of multimoding in solid state lasers has been attributed to the lack of fast spatial cross relaxation[16-17] and to the lack of adequate discrimination among the modes[18]. In their steady-state theory of quantum oscillators in a multimode cavity, Wagner and Birnbaum[18] have shown that very intense line narrowing occurs in a multimode resonator when only a small fraction of the modes participate in the oscillation, and when the initial pump is above a certain level. It is not at all certain, however, that the enormous linewidth narrowing would also occur in giant pulse operation, for which a steady state never exists. Fleck[19] has considered the linewidth problem in some detail, noting that there exist two limiting cases. In one case when the laser operates in a single mode in the sense of Fox and Li[20],

linewidth narrowing is expected to occur, and is correctly given by the Schawlow-Townes formula[21]. In the other case when the number of back and forth reflections of the amplified spontaneous radiation within the cavity is not sufficient to establish a single mode, the narrowing is expected to be only moderate. This idea has been incorporated in the phenominological description of Sooy[22], who related the mode selection to the mode buildup rates in a passive Q-switched laser. We shall show in this paper that the mode structure of the giast pulse laser follows directly from the rate equations, and that the output linewidth is essentially determined by the effective number of reflections within the cavity and by the gain (or loss) profile of the laser.

THE RATE EQUATIONS

Following Hellwarth[3], we neglect the effects of continued pumping and of spontaneous emission during the entire span of the giant pulse. We assume that the cavity longitudinal modes are symmetrically spaced with respect to the center of the molecular resonance, with the most favored mode right at the center. At frequencies not too far off from the center, the absorption coefficient in the unexcited state is essentially frequency-independent, and will be designated α_o. For sake of simplicity, we shall ignore the presence of off-axial transverse modes, although in practice each longitudinal mode is usually accompanied by a number of such transverse modes.

The laser is characterized by the following parameters: ℓ, the length of the laser material, and L, the optical distance between the end reflectors of the cavity. It follows that the time of a single passage through the cavity is $t_1 = L/c$, where c is the speed of light, and that the lifetime of a photon in the ith mode is $T_i = t_1/\gamma_i$, where γ_i is the loss coefficient expressing the fractional photon loss per pass in the ith mode due to reflection and incidental losses. In particular, the most favored mode will be designated the zeroth mode, and thus the corresponding loss coefficient will be γ_o, and the corresponding photon lifetime $T_o = t_1/\gamma_o$.

Under the aforementioned conditions, it can be shown[5, 16] that the behavior of the giant pulse in the period following the switching is governed by the following set of nonlinear differential equations:

$$\frac{dn}{dt} = - \sum_i (n/n_p)\, \phi_i \tag{1}$$

$$\frac{d\phi_i}{dt} = \left[(n/n_p) - b_i\right] \phi_i \tag{2}$$

where the summation is carried over all the longitudinal modes excited above threshold. Here, n is the ratio of the population inversion per unit volume to the density of the active ions N_o in the laser crystal; ϕ_i is the ratio of the photon density in the ith mode to $N_o/2$; and t is the time in units of the lifetime T_o of the photons in the zeroth mode in the cavity. Other relevant quantities are

$$\alpha_o \ell\, n_p = \gamma_o \tag{3}$$

and

$$b_i = \gamma_i/\gamma_o \tag{4}$$

The temporal behavior of a multimode giant pulse laser is much the same as that described by the single-mode theory, as may be apparent upon examination of Equations (1) and (2). It is of interest to compare the total energy obtainable from the pulse under single-mode and multimode operations. To this end, Equations (1) and (2) may be integrated and combined to show that

$$\sum_i b_i \int_o^\infty \phi_i\, dt = n_o \left\{ 1 - \exp\left[-(1/n_p) \sum_i \int_o^\infty \phi_i\, dt\right]\right\} \tag{5}$$

where n_o is the initial value of n. If one further writes

$$\sum_i b_i \int_o^\infty \phi_i \, dt = b \sum_i \int_o^\infty \phi_i \, dt \tag{6}$$

so that b is the average loss coefficient over the participating modes, Equations (5) may be transformed to become

$$(b\, n_p) \ln (n_f/n_o) = n_f - n_o \tag{7}$$

where n_f is the final value of n. Thus, the total energy obtainable from a multimode pulse is the same as that from a single-mode pulse having a loss coefficient of $b\,\gamma_o$. Such a result has been implied in the work or Wagner and Birnbaum[18]

It has been assumed that there exists a longitudinal mode which has the lowest loss rate and which is exactly on the molecular resonance. Such conditions may be realized through use of transmission or reflection mode selectors[11]. It will be shown later that within a transmission band $\Delta\nu_1$ of the mode selector, the normalized loss coefficient b_i is of the form

$$b_i \equiv f(\Delta\nu) = 1 + (1/\gamma_o) \ln \left[1 + (\Delta\nu/\Delta\nu_1)^2\right] \tag{8}$$

where $\Delta\nu$ is the frequency deviation from the most favored mode. It is sufficient for our purpose to write

$$\Delta b_i = b_i - b_o \tag{9}$$

and assume that the initial photon density is the same in all participating modes. We then obtain from Equation (1)

$$\int_o^\infty \phi_i \, dt = (1/b_i) \int_o^\infty n \, \phi_i \, dt = \int_o^\infty e^{-\Delta b_i t} \phi_o \, dt \tag{10}$$

$$\approx e^{-\Delta b_i t_m} \int_o^\infty \phi_o \, dt \tag{11}$$

where the last form follows from the fact that the principal contribution to the integral comes from a small region centered about t_m when the photon density in the most favored mode peaks. t_m is thus the time from switching to the occurrence of the peak, and may to a good approximation be given by[5]

$$t_m = \frac{1}{[(n/n_p) - 1]} \ln \left[1 + \frac{(n_o - n_p)}{N_e \phi_{oo}} \right] \tag{12}$$

where ϕ_{oo} is the normalized initial photon density, and N_e is the effective number of participating modes. Because of the nature of the logarithmic dependence, the result is seen to be insensitive to both N_e and ϕ_{oo}.

The linewidth $\Delta\nu_o$ of the output as measured between half-maximum intensity points is obtained from the relation

$$\int_o^\infty \phi_i \, dt \Big/ \int_o^\infty \phi_o \, dt = e^{-\ln 2} \tag{13}$$

Substitution of Equations (8) and (11) in Equation (13) gives

$$\Delta\nu_o = \left[2^{\gamma_o/t_m} - 1 \right]^{\frac{1}{2}} \Delta\nu_1 \tag{14}$$

It is important to note that t_m/γ_o is equal to the number of single passage transits which the spontaneous radiation has to make before it reaches the peak density. Equation (14) states that the output linewidth narrows as the number of transits through the cavity is increased. At high initial pump levels, examination of Equations (12) and (14) further shows that the giant pulse builds up from spontaneous noise too quickly to allow appreciable narrowing in the output. Although the above considerations are formulated in terms of the loss profile of Equation (8) in the cavity, similar linewidth narrowing is also expected to occur for a frequency-dependent gain profile in the cavity.

TRANSMISSION MODE SELECTION

Our consideration of total emitted energy in the pulse is in general agreement with experimental results reported to date. McClung and Weiner[11] observed that the power from a single-mode pulse is approximately the same as that from a multimode pulse.

To test the theory in regard to the narrowing of the output linewidth, we have performed experiments with a transmission mode selector inserted in the cavity. Such a transmission mode selector provided the necessary discrimination among the longitudinal modes. The experimental arrangement is depicted schematically in Figure 1. The laser consisted of an ultra high quality Linde ruby rod with 60° orientation, ¼" in diameter, 3" in length, and Brewster-angle cut at one end. The uncoated flat end of the ruby served as one end of the cavity, and the other end of the cavity was provided by a motor driven roof prism rotating at 24,000 rpm. The optical length of the cavity was 67 cm, and the corresponding spacing between adjacent longitudinal modes was 0.004 cm^{-1}. The ruby was cooled to $-80^{\circ}C$ by flowing cold nitrogen gas, and self lasing was prevented because of the nature of the Brewster-angle cut. A sapphire mode selector ¼" thick was placed a few centimeters in front of the rotating prism, and was tilted about an axis parallel to the prism refracting edge to obtain optimum performance. Optical flats of varying thickness and with coated surfaces to provide higher surface reflectivity were also used as mode selectors.

The intensity transmissivity $\underline{T}$ of the sapphire flat is given approximately by

$$\underline{T} \approx \frac{1}{1 + (\Delta\nu)^2 \, (\pi \underline{F}_s/\Delta\nu_s)^2} \qquad (15)$$

where $\Delta\nu$ is the frequency deviation from the center of the transmission band, $\underline{F}_s = 0.6$ is the finesse, and $\Delta\nu_s = 0.45\ cm^{-1}$ is the mode spacing of the sapphire flat. Similarly, the prism reflectivity $\underline{R}_2$ is given by

$$\underline{R}_2 \approx \frac{1}{1 + (\Delta\nu)^2\ (\pi \underline{F}_p/\Delta\nu_p)^2} \tag{16}$$

For the quartz prism used, $\underline{F}_p = 0.385$ and $\Delta\nu_p = 0.27\ cm^{-1}$. It is seen that the Fabry-Perot cavity formed by the ruby, the roof prism and the sapphire flat is characterized by a frequency-dependent loss coefficien. $\gamma(\Delta\nu)$

$$\gamma(\Delta\nu) = -\tfrac{1}{2} \ln (\underline{R}_1\ \underline{R}_2\ \underline{T}^2) \tag{17}$$

where $\underline{R}_1 = 0.07$ is the reflectivity at the ruby-air interface. Equation (8) then follows by identifying

$$\gamma_o = -\tfrac{1}{2} \ln \underline{R}_1 \tag{18}$$

and

$$(\Delta\nu_1) = 0.8(\Delta\nu_s) \tag{19}$$

The output linewidth was observed with a Fabry-Perot interferometer. Tne instrument linewidth was about $0.01\ cm^{-1}$. When the sapphire mode selector was removed from the cavity, the output was found to contain two components each $0.1\ cm^{-1}$ wide and spaced $0.27\ cm^{-1}$ from each other; this corresponded to the mode spacing of the roof prism. At higher pump levels, up to five such components were observed. With the mode selector inserted in the cavity, the output was found to contain only one component approximately $0.04\ cm^{-1}$ in width (Figure 2a), which is amply above the instrument linewidth; occasionally a weak component spaced $0.45\ cm^{-1}$ away from the firts component was also observed (Figure 2b).

To observe such spectral characteristics, and avoid multiple spiking in time, it was found necessary to operate the laser barely above threshold.

Taking $n_p = 0.10$, $n_o = 0.125$, $\gamma_o = 1.28$ and $N_e\phi_{oo} \approx 5 \times 10^{-16}$, we found from Equation (14) that

$$\Delta\nu_o \approx 0.0665\ (\Delta\nu_s) = 0.03\ \text{cm}^{-1}$$

This agrees with the experimental value. Subsequent experiments using dielectric coated optical flats yielded slight improvement in output linewidth; similar agreement was also observed between the experimental and the calculated values.

The above considerations have been given for longitudinal modes; similar considerations should also apply for the off-axial transverse modes when there exists discrimination among these modes. Although transmission mode selection alone is insufficient to warrant operation in a single longitudinal and transverse mode, it is gratifying to note that our analysis serves to show that such a single-mode operation with a rotating prism giant pulse laser should indeed be possible, (as has already been observed) provided that adequate discrimination among the longitudinal as well as transverse modes prevails, and that the operation is carried out at a pump level not too far above the threshold.

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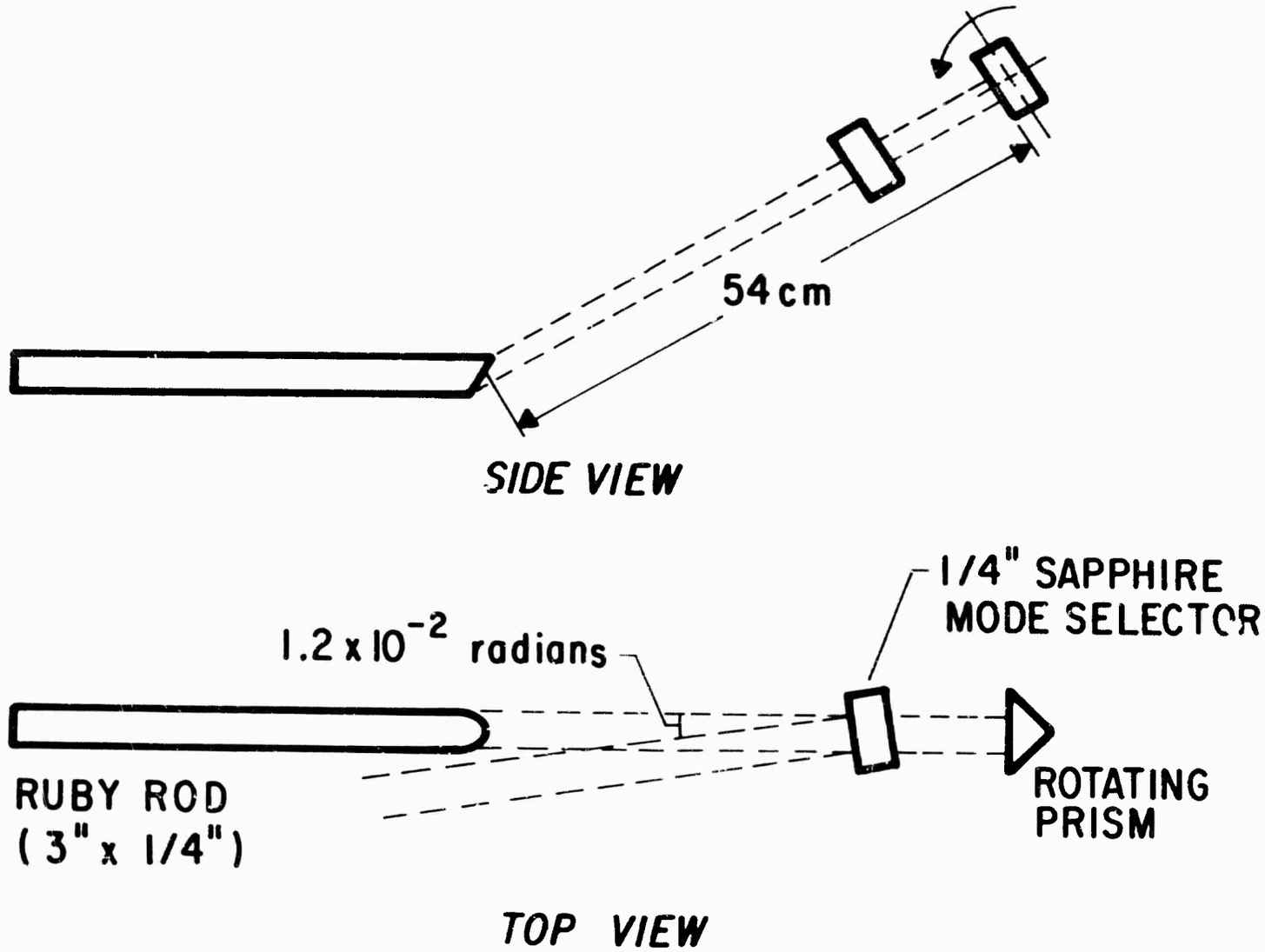
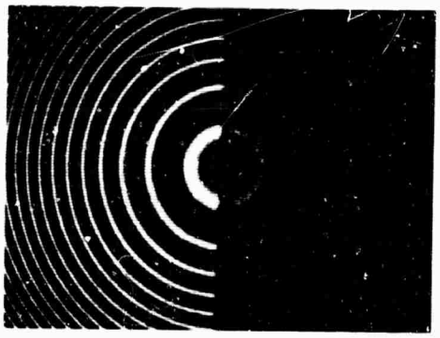
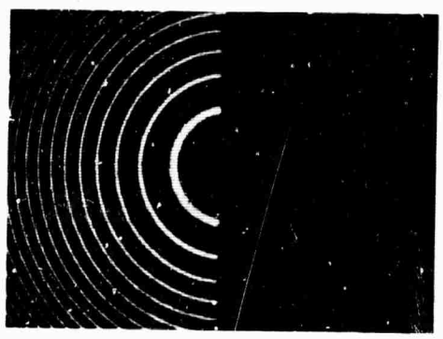


Schematic of the experimental setup

FIGURE 1



(a)



(b)

Fabry-Perot patterns showing the spectral characteristics of the giant pulse obtained with transmission mode selection. The interorder spacing of the Fabry-Perot etalon was 0.33 cm^{-1}, and a Corning 1-58 glass filter covers half of the patterns.

FIGURE 2

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13. ABSTRACT

This report summarizes work done at Philco Applied Research Laboratory under contract Nonr-4850(00). Extensive measurements were taken on the conversion efficiency and threshold of stimulated Raman emission in benzene, nitrobenzene, toluene, and carbon disulfide. It has been shown that the self-focusing action of a laser beam is responsible for the onset of stimulated Raman emission.

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