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Semi-Annual Report

OPTICAL SWITCHING BY STIMULATED THERMAL RAYLEIGH SCATTERING

L.M. PETERSON
Optical Science Laboratory
Advanced Concepts Division
JUNE 1986

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ABSTRACT

Preliminary experiments were conducted whose ultimate goal is to develop all-optical control functions useful in an all-optical or optical-electronic hybrid digital computer or for optical interconnects. Stimulated thermal Rayleigh scattering (STRS) based upon generator experiments was pursued for scattering angles of 90° and 180° (backscattering). A pulsed nitrogen laser pumped dye laser served as the radiation source and the interaction medium was a liquid to which an absorbing dye was added.

STRS amplifier experiments were successful and gain was observed and studied parametrically using eosine dye in ethanol. The gain was found to increase (although the gain coefficient decreased) with increasing pump power and the gain was found to be a maximum at an absorption coefficient of about 2.6 cm⁻¹. The generator experiments did not lead to stimulated scattering due to the limited output power of the laser and its multi-longitudinal spectral mode content. These studies will be continued along with analytical modeling in order to characterize the interaction and to enable the optimization of the scattering process.

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 A circled "deg" is written above the word "generator" in the first paragraph.
 Another circled "deg" is written to the right of the word "90°" in the first paragraph.
 A circled "720 cm" is written below the word "cm⁻¹" in the second paragraph.



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PREFACE

The work reported here was performed in the Optical Sciences Laboratory, which is part of the Advanced Concepts Division at ERIM. The work was supported by the Defense Advanced Research Projects Agency under the direction of Dr. John A. Neff and was monitored by the Air Force Office of Scientific Research under Contract No. F49620-84-C-0067. Col. Robert Carter was program manager for the AFOSR during the initial phases of this program and was replaced by Dr. Lee Giles later on in the program.

This semi-annual technical report covers work performed from 15 June 1984 to 15 February 1985. The principal investigator at ERIM was Dr. Lauren M. Peterson and Patrick Hamilton was a major contributor to the experimental work.



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OPTICAL SWITCHING BY STIMULATED THERMAL
RAYLEIGH SCATTERING

1
INTRODUCTION AND SUMMARY

During the first experimental phases, attempts were made to explore stimulated thermal Rayleigh scattering (STRS) based upon generator experiments. Conventional 180° backscattered STRS [1-4] was pursued using usual spherical optics and also the less conventional 90° scattering which was proposed by us using cylindrical optics. Neither effects were observed, probably due to the limited power output of our pulsed dye laser source. Significant power is required for generator experiments since the source of the STRS is only spontaneous Rayleigh noise. Also, the multi-longitudinal mode output of the dye laser may have been an additional deterrent.

The generator experiments were therefore temporarily abandoned in favor of more basic and controllable amplifier experiments. Here, instead of a signal which originates from spontaneous noise, a signal of chosen magnitude, direction, and frequency is injected and gain is measured parametrically.

Using spherical optics and a single beam of focused pump radiation, the gain favors scattering at the larger angles. Since the interaction region is longest along the optic axis, 180° backscattering is maximum. Gain experiments were therefore conducted with a forward traveling pump and a backward traveling probe beam whose frequency could be shifted using an acousto-optic modulator. Gain was observed using eosine dye in ethanol. The gain was found to increase with increasing pump power while the gain coefficient was found to decrease with increasing pump power. Gain was found to maximize at about 0.08 cm/Mw for an absorption coefficient of about 2.6 cm^{-1} .

2
GENERATOR EXPERIMENT

Experiments were attempted to observe 90° STRS (Appendix A-3) in absorbing liquids using cylindrical optics as shown in Figure 2-1. Radiation from our Molecron SP-10 nitrogen laser pumped dye laser (Table I and Figure 2-2) was weakly focused in one dimension using an 80 mm focal length cylindrical lens, and tightly focused using a 25 mm or 18 mm focal length microscope objective. The presence of the cylindrical lens results in two orthogonal lines of focus to be formed. A glass cuvette containing a liquid sample was placed so that the first and horizontal line focus was situated at its center. An f/2 camera lens was used to gather radiation emanating from this line focus and direct this radiation to an RCA 1P28 photomultiplier tube. Appropriate baffling was required to prevent light scattered by the cuvette walls from getting to the PM tube.

The Molecron SP-10 laser was operated using coumarine 500 dye with output at $0.5 \mu\text{m}$ and rhodamine 6 G at $0.59 \mu\text{m}$ to provide pump radiation. A sample cuvette filled with water to which a drop of milk was added as a scatterer facilitated the alignment process. Methanol, carbon disulfide, and carbon tetrachloride solvents were used along with iodine as an absorber for observing STRS. Although several solvents and numerous dye concentrations (absorptivities from 0.3 to 2.0 cm^{-1}) were used, no significant scattering attributable to STRS was observed.

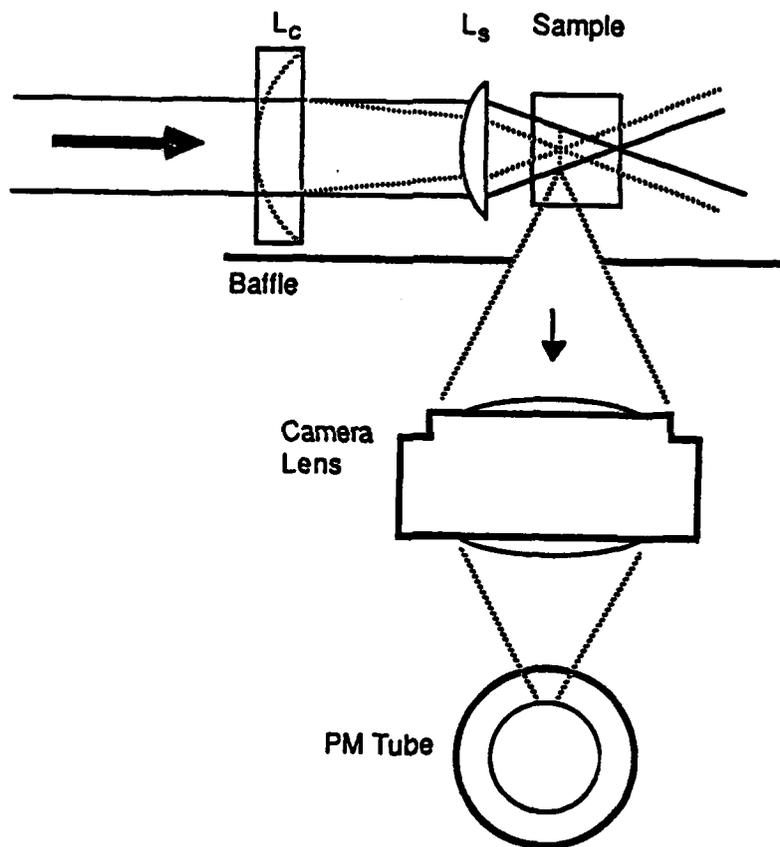


Figure 2-1 Generator Experiment. The dotted lines represent rays in a plane orthogonal to that of the figure resulting in a line focus within the sample.

TABLE I
MOLECTRON SP-10 LASER SPECIFICATIONS

N₂ Laser

| | |
|------------------|---------------------------|
| Wavelength | 337.1 nm |
| Energy per Pulse | 0.5 mJ |
| Pulse Width | 9 ± 0.5 nsec |
| Peak Power | 50 KW |
| Beam Size | 1.5 x 25 mm |
| Beam Divergence | 7 x 0.8 mrad (half angle) |
| Radiation Rate | 0 - 120 pps |
| Gas Pressure | 25 torr |

Dye Laser*

| | |
|-------------------|---------------------------------------|
| Wavelength Range | 360 - 740 nm |
| Line Width (FWHM) | 0.3 nm, 12 cm ⁻¹ , 360 GHz |
| Pulse Width | 7 nsec |
| Beam Diameter | 0.6 mm (nominal) |
| Cavity Length | 12 cm |

| | <u>Coumarin 500</u> (500 nm) | <u>Rhodamine 6 G</u> (590 nm) |
|------------------|---------------------------------|----------------------------------|
| Peak Power | 10 Kw | 5 Kw |
| Energy per Pulse | 70 μJ | 35 μJ |

*Dye cuvette with stirrer; Littrow grating.

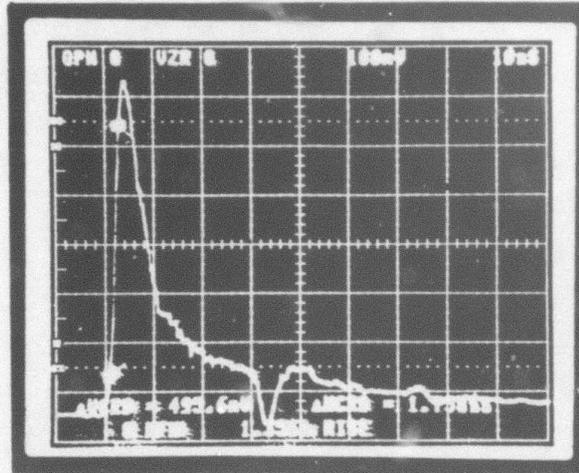


Figure 2-2 Oscilloscope trace of dye laser pulse. The risetime is limited by that of the 300MHz preamp. (Note the cable reflection at 30 nsec.) 10 nsec/division.

3
GAIN EXPERIMENTS

Generator experiments in stimulated scattering rely upon spontaneous scattering or spontaneous noise as the initiating signal which is amplified to levels approaching the magnitude of the pump laser itself. Since the spontaneous scattered radiation is extremely weak, total gains on the order of e^{30} , 10^{13} or 130 dB are required. (Spontaneous scattering has an efficiency of about 10^{-6} and is omnidirectional, scattering the light into a full 4π steradians. The laser and stimulated scattering, however, are unidirectional with a divergence of only about 10^{-3} radians or a solid angle of only 10^{-6} steradian. Spontaneous scattering therefore provides only $10^{-6} \times 10^{-6} \div 4\pi$ or about 10^{-13} of the incident radiation to initiate the stimulated scattering process.) If however, a modest signal at the control of the experimenter, is injected with the proper characteristics, amplification or gains as small as $e^{0.1}$ or 0.5 dB can be measured.

3.1 EXPERIMENTAL ARRANGEMENT

First attempts were made to obtain gain in the 90° , transverse geometry of Figure 2-1, but this was quickly abandoned due to the difficulty of injecting a signal beam along the transverse focal volume whose dimensions were only about $15 \times 500 \times 500 \mu\text{m}$. The longitudinal geometry of Figure 3-1 was adopted and uses strictly spherical optics such that gain is measured in the backward direction.

Radiation from the Molelectron SP-10 nitrogen laser pumped dye laser (5.5 kw, $44 \mu\text{J}$, 7 nsec at $0.51 \mu\text{m}$) was focused into the 1 cm square sample cuvette using a 48 mm focal length microscope objective lens L2. A variable neutral density filter, N, was used to control the power level of this pump radiation, and a beamsplitter, S_2 , which preceded the focusing lens was used to sample the radiation

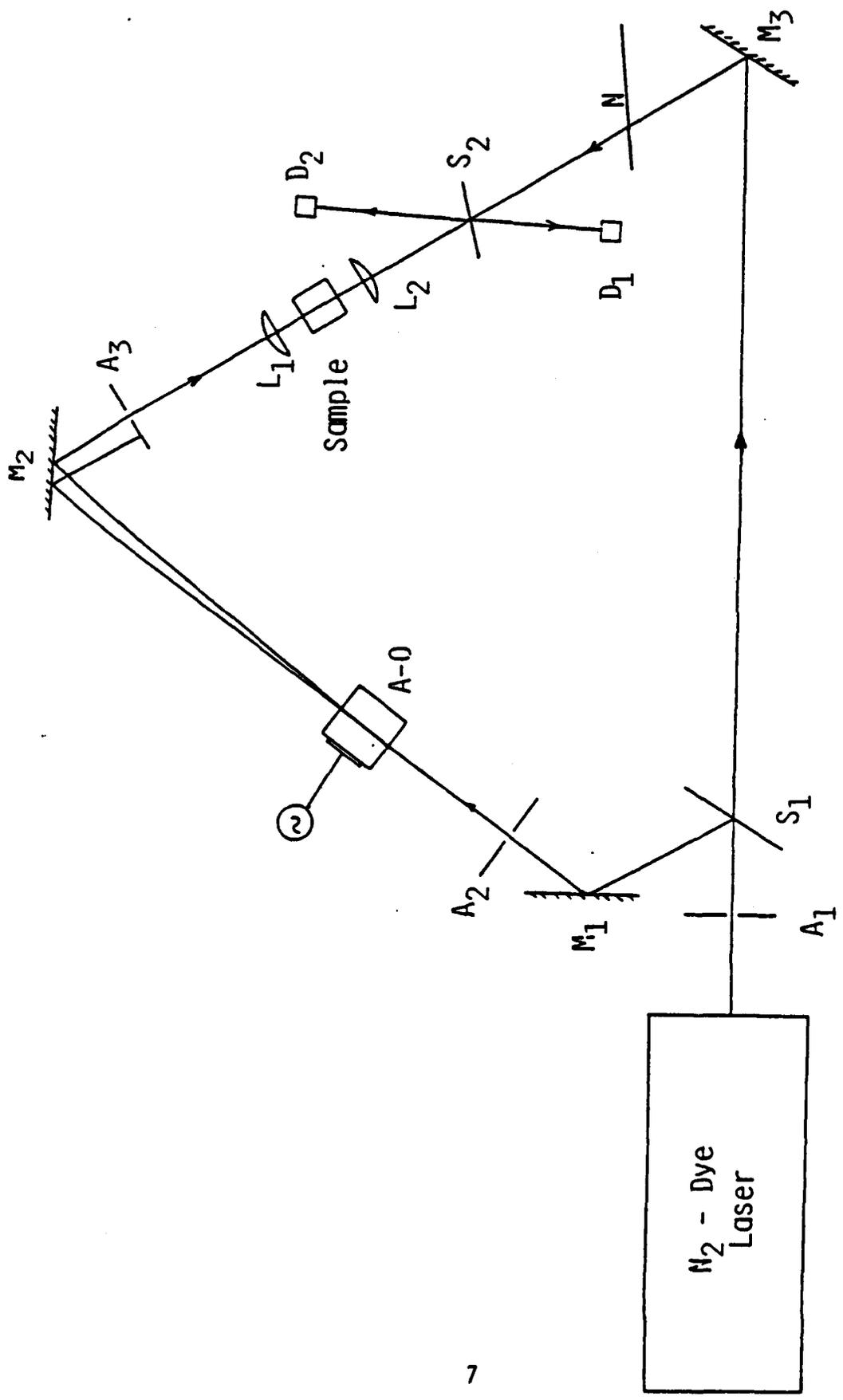


Figure 3-1 Experimental arrangement for measuring gain due to STRS.

and direct it to a high-speed PIN (HP 4203) photodiode, D_1 . Roughly 2 percent of the pump radiation was directed, using an uncoated microflat plate beamsplitter, S_1 , along a separate path to the sample cell. This radiation could be up- or down-shifted in frequency from 30 to 50 MHz (or possibly multiples of these frequencies if higher orders were used) using an acousto-optical cell before being focused into the sample cuvette using another 48 mm focal length microscope objective lens L_1 . With care, the two opposing beams could be made precisely counterpropagating but precise overlap of the two beams at their 30 μm diameter focus was difficult. It was found that the power density ($\sim 10^9 \text{w/cm}^2$) of the focused pump beam was sufficient to burn a small hole through a piece of metal foil held in place at the center of the cuvette within the liquid sample. Transverse displacement of the probing signal beam by transverse alignment of the focusing lens optimized the transmission of the probe radiation through the burned aperture. The same beamsplitter, S_2 , which sampled the incident pump radiation was used to sample the transmitted probe radiation as shown in the figure. Gain was indicated if the signal at this second detector was greater in the presence of the pump radiation than in its absence.

3.2 GAIN MEASUREMENTS

Measurements were made using the experimental configuration, shown in Figure 3-1, with the Molelectron SP-10 nitrogen laser pumped dye laser. Approximately 0.1 percent or 5 watts of peak power was focused into the sample cuvette using lens L_1 , and the signal level was measured using detector D_2 in the absence of any pump radiation. In the presence of pump radiation (variable up to 5 KW), focused into the cuvette by lens L_2 , an increase in signal at detector D_2 is indicative of gain, or amplification of the probe beam. (Pump light backscattered from lens L_2 , the sample cuvette or any other optics was determined by blocking the probe beam and

measuring the radiation at D_2 . This level was subtracted from the measurements at D_2 in determining the gain.)

Gain measurements were attempted for several solvent-dye combinations including CS_2 , CCl_4 , CH_3OH and C_2H_5OH with I_2 or eosine. Significant gain was measured using eosine dye in ethanol, absorption coefficients of about 0.9 cm^{-1} at 500 nm and 2.6 cm^{-1} at 52.5 nm. Figure 3-2 shows how the gain ($\ln \tau$) and gain coefficient (g) vary as a function of the incident pump power level for the laser tuned to the two different wavelengths. The nonlinear gain increases with increasing pump power while the gain coefficient decreases with pump power. We assume that the gain mechanism is due to the beating between the strong pump radiation and the weak probe radiation to create a standing wave fringe pattern in the sample. Through absorption and thermalization, a standing wave phase grating is formed, which is able to diffract (Bragg reflect) the incoming pump radiation. The amplified probe radiation is actually reflected copropagating pump radiation.

Phase matching of the Bragg reflected pump radiation with the probe radiation should, according to STRS theory [1, 3, 4], require a slightly higher frequency for the probe beam relative to the pump beam (a slightly lower frequency should lead to Bragg reflection of the probe radiation, i.e., loss for the probe beam and amplification for the pump beam). We observed no difference in gain between unshifted probe radiation and probe radiation which was either up- or down-shifted by 40 MHz in our experiments.

The gain coefficients in Figure 3-2 are significantly less than the 0.2 cm/Mw which is considered typical for STRS [2-4]. These low values and our inability to generate STRS from spontaneous noise [Section 2] may be due to the multimode nature of the dye laser output. From Table I, according to the manufacturers specifications, the laser linewidth is about 360 GHz (or more) whereas the separation

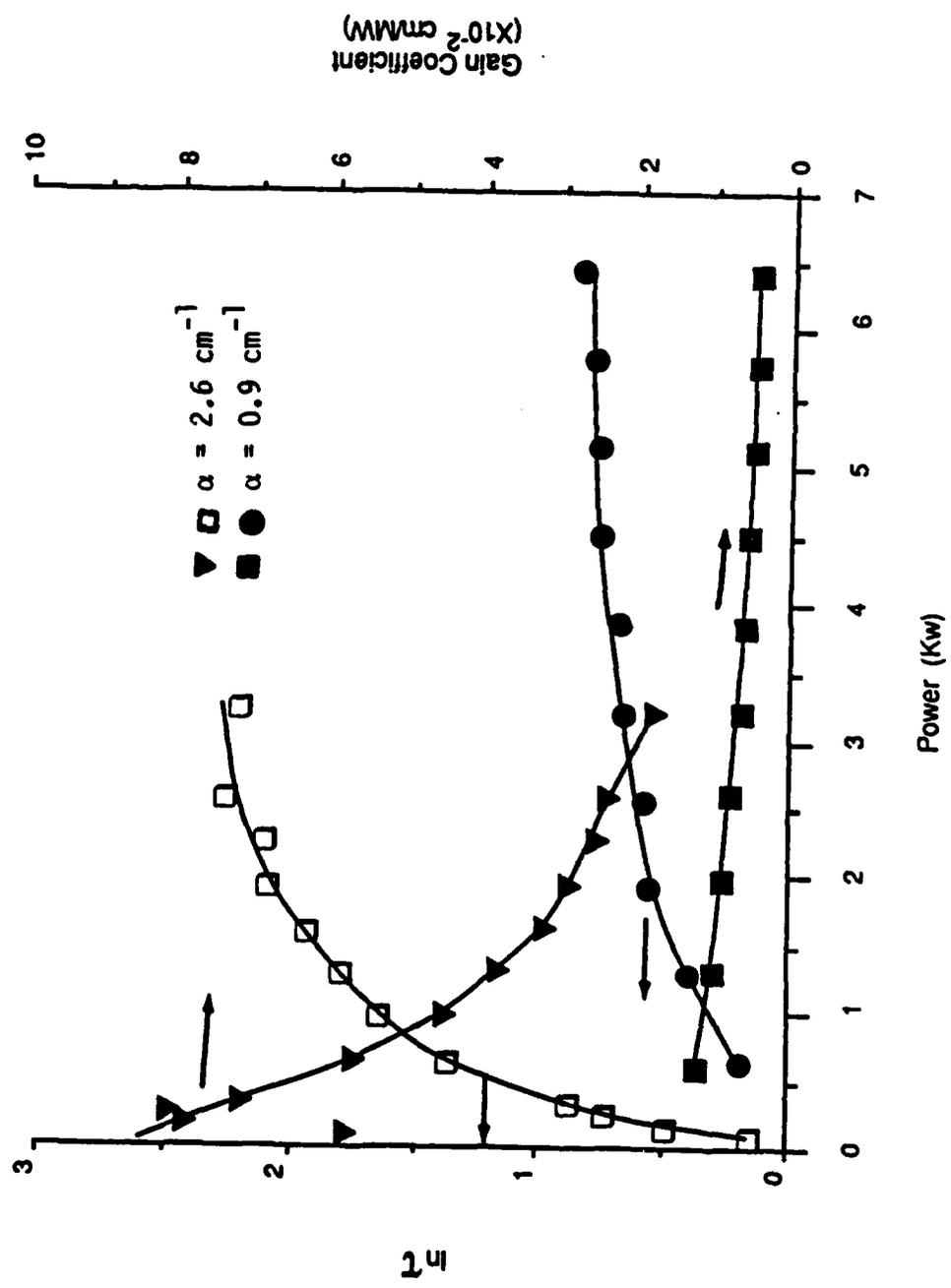


Figure 3-2. Measured gain (Int) and gain coefficient for eosine dye in ethanol.

between laser resonator modes ($c/2L$) is 2.5 GHz leading us to expect about 144 modes oscillating simultaneously. Since the average laser power per mode is the total power divided by the number of modes, the gain coefficient per mode may therefore be increased by a factor equal to the number of modes. Or, examining the theoretical equation for the gain [1,3,4], a broadband laser can be expected to have a gain coefficient which is reduced by a factor of Γ_L/Γ_R (where Γ_L and Γ_R are laser and Rayleigh linewidths respectively) over that for a narrow ($\Gamma_L \ll \Gamma_R$) laser linewidth.

4
SUMMARY

Preliminary experiments were conducted which examined stimulated thermal Rayleigh scattering as a mechanism for achieving optical control functions. Generator experiments for producing high intensity directional scattered radiation which originates from amplified spontaneous noise radiation requiring gain on the order of e^{30} was not observed with our pulsed nitrogen laser pumped dye laser.

Amplifier experiments were successful and gains on the order of e^2 were observed for eosine dye in ethanol with an absorption coefficient of about 2.6 cm^{-1} . The greatest gain coefficient which we measured was 0.08 cm/Mw and is within a factor of 3 of what is considered typical for STRS [3].

The inability to observe STRS which is amplified from spontaneous noise and the low gain observed in the amplifier experiments is likely attributable to the spectrally broad, multi-longitudinal mode laser output of our dye laser. The use of a spectrally narrower laser, either by modification of our existing laser or by purchasing a replacement laser, will be pursued in subsequent phases of this program.

APPENDIX A-1 PHASE MATCHING

Any gain imparted to the probing signal radiation is presumably a consequence of the beating between the signal radiation and the pump radiation which leads to a standing wave or fringe pattern in the interaction medium. If the probe radiation is exactly the same frequency as the pump radiation, then the fringe pattern is stationary in space. If the probe radiation frequency is different from the pump frequency, then the fringe pattern moves longitudinally. This motion may serve to provide phase matching in the two-wave mixing process.

The interference pattern created by the interaction of the counter-propagating beams leads to a thermal fringe pattern due to the absorption by the dye present in the liquid sample. This thermal fringe pattern leads to a refractive index fringe pattern or phase grating according to the magnitude of dn/dT for the liquid solvent ($-dn/dT \sim 10^{-4} \text{K}^{-1}$ for liquids [5] and 10^{-5}K^{-1} for solids [6]). Since the standing wave spatial periodicity is $\lambda_0/2n$, the Bragg condition for 180° backscattering of the pump radiation is satisfied. What appears as gain of the probe signal radiation is actually pump radiation which has been redirected (reflected) by Bragg scattering to propagate with the transmitted signal radiation.

If the path length difference between the pump beam and the probe beam, measured from the beamsplitter to the focal region in the cuvette is greater than the coherence length of the laser, then the instantaneous frequency of the pump and probe beams will be different enough such that the interference fringes will move back and forth in time such that the time averaged pattern is washed out and little or no gain can be expected. If, however, the path lengths are equal to within a coherence length, and the fringes can be made to move at a slow, constant velocity, then the gain might be enhanced due to

enhanced phase matching between the scattered pump radiation and the transmitted probe radiation. The velocity, v , for enhanced phase matching would approximately correspond to motion of a half-wavelength in the time corresponding to the thermal relaxation time, i.e., $v = \lambda/2\tau$. This fringe motion can be established by shifting the frequency of the probe radiation by an amount

$$\Delta\nu = 2 \frac{v}{c} \nu = 2 \frac{v}{\lambda}$$

$$\Delta\nu = \frac{1}{\tau}$$

Since thermal relaxation times are on the order of 10^{-8} sec [7], this would correspond to frequency offsets on the order of 100 MHz. This is in the range of acousto-optic beam deflectors. We note that τ is of the same order of magnitude as the duration of the laser pulse such that the fringe pattern moves a fraction of a fringe or a few fringes at the most during the time of the pulse.

APPENDIX A-2
GAIN COMPUTATIONS

The gain of the pump-probe interaction in the liquid-dye sample was determined using measurements depicted in Figure A2-1. Pump radiation incident from the left in Figure A2-1(a) interacts with the signal or probe radiation incident from the right to provide amplification or gain to that probe radiation. The amplified radiation intensity is detected using the beamsplitter, BS, and the detector shown, and is represented by a measurement V_a . The probe or signal radiation in the absence of gain is measured as V_p in Figure A2-1(b) and is attained by simply blocking the incident pump radiation prior to the beamsplitter. Since the pump radiation is much stronger than the probe radiation, background radiation due to scattering or reflections from the beamsplitter, lenses, windows or the liquid sample, may be significant. This background is measured by V_b , shown in Figure A2-1(c) by simply blocking the incident probe radiation.

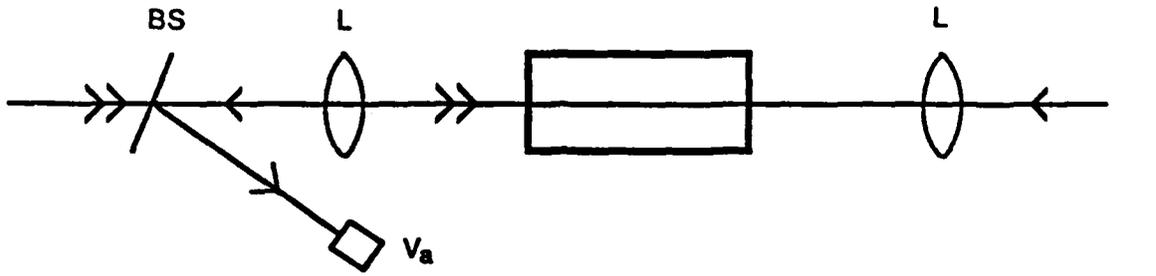
The gain is assumed to be exponential, such that the transmission of the sample in the presence of pump radiation is

$$T = e^{gIl} = \frac{I_{out}}{I_{in}} = \frac{V_a - V_b}{V_p}$$

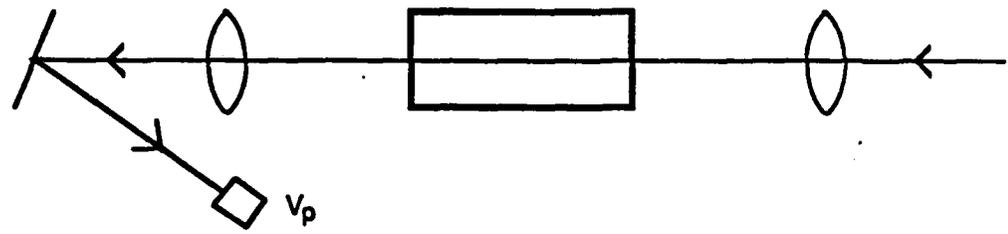
The gain coefficient g is computed as

$$g = \frac{\ln \left(\frac{V_a - V_b}{V_p} \right)}{Il}$$

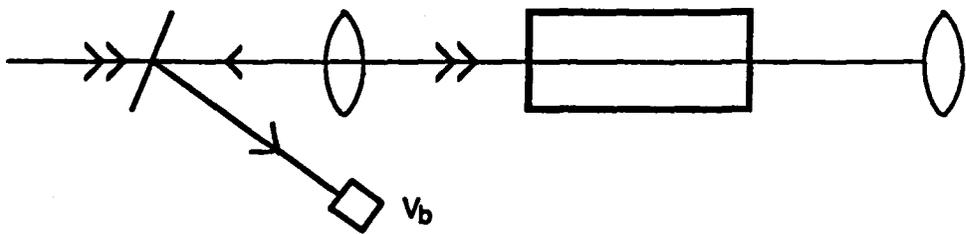
The I in the denominator is the intensity of the pumping radiation multiplied by the interaction length l in the focal region where the stimulated scattering and the gain occur. (This might be considered as a special case of forced Rayleigh scattering [8] where $\theta = 180^\circ$ and the pump radiation is its own degenerate probe.) The pump



(a) Signal Radiation Gain



(b) Signal Alone



(c) Scattered Pump Radiation

Figure A2-1 Gain Measurements. Pump radiation is designated by a double arrow; probe and stray radiation are designated by a single arrow.

intensity in the focal region in watts cm^{-2} is simply the raw laser power, P_L , divided by the area of the waist whose diameter is δ . The length, l , of the focal region where the intensity is highest and where the nonlinear optical interaction occurs is defined as the confocal parameter [9] or the Rayleigh range [10]

$$l = \frac{\pi}{4} \frac{\delta^2}{\lambda} .$$

Therefore, we have

$$\begin{aligned} I l &= \frac{P_L}{\frac{\pi}{4} \delta^2} \cdot \frac{\pi}{4} \frac{\delta^2}{\lambda} \\ &= \frac{P_L}{\lambda} \end{aligned}$$

To first order, I is independent of focal spot size or interaction length and therefore is independent of the choice of focal length or f-number of focusing optic. We may write our final equation for the gain coefficient as

$$g = \lambda \frac{\ln \left(\frac{V_a - V_b}{V_p} \right)}{P_L} \quad \frac{\text{cm}}{\text{watt}}$$

APPENDIX A-3
STIMULATED SCATTERING AT 90°

Stimulated scattering is commonly observed in the backward (180°) direction not because the gain is greatest for this direction but because of the geometry of the propagating beam of radiation. The nonlinear amplification depends upon the field intensity of the radiation and upon the nonlinear susceptibility of the material medium. In the focal region of a spherical lens as shown in Figure A3-1, the field intensity is high and the coefficient, g , for SRS and STS is roughly the same in all but the forward direction. (More precisely, a $\sin \theta/2$ distribution is encountered.) Although the gain coefficient for a given medium is roughly constant, the amplification process is exponential as

$$I_s(l) = I_s(0) e^{I_L l}$$

where I_L is the laser intensity I_s is the stimulated scattered radiation and l is the interaction length. Although there is amplification of radiation in all directions in Figure A3-1, l is very small in all except the forward and backward directions. For such a geometry, since g is zero in the forward direction, this leaves stimulated scattering in the backward direction i.e., for $\theta = 180^\circ$. (We note that the gain coefficient for SRS is high in the forward direction also and as such, SRS is observed in both the forward and backward directions using the geometry of Figure A3-1.)

If we consider a material medium in Figure A3-2 to be the same as that in Figure A3-1, then the gain coefficient, g , should be the same. Since a cylindrical rather than a spherical lens is used however, the focal volume is now an elongated region which is oriented transverse to the direction of propagation of the radiation. Since l is longest for a scattering angle, θ , of 90° amplification can be expected to be greatest in this direction.

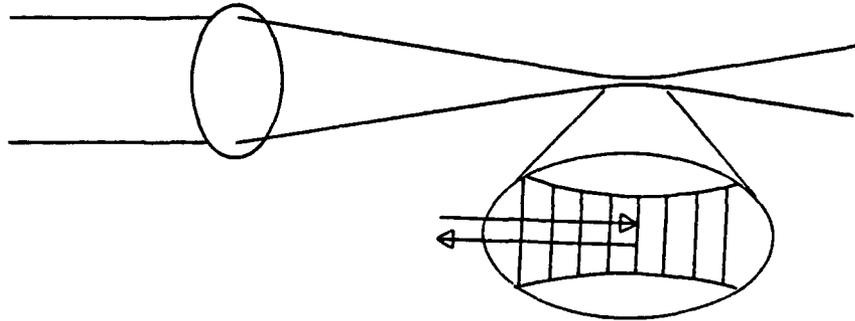


Figure A3-1 Longitudinal focal geometry using a spherical lens

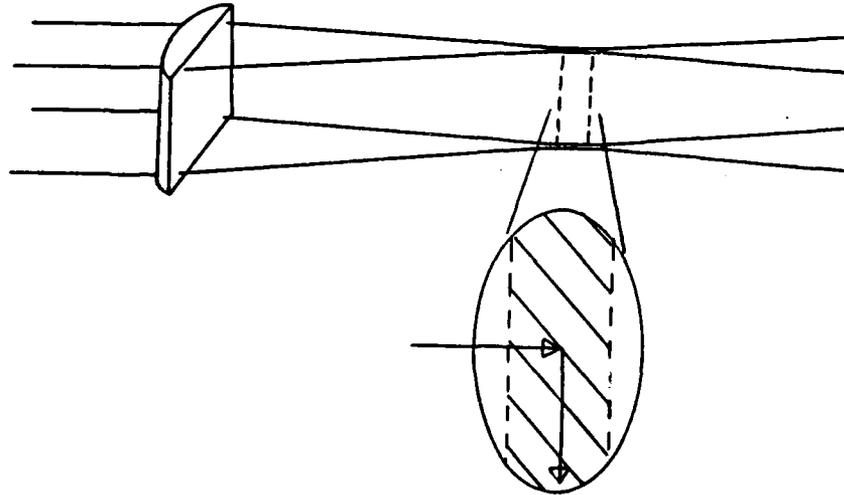


Figure A3-2 Transverse focal geometry using a cylindrical lens

Examining this gain process on a microscopic scale, Figure A3-1 can be used to visualize the interference between backscattered radiation and forward propagating laser radiation to produce interference fringes whose iso-density planes are perpendicular to the laser propagation. The planes are separated by $\lambda_0/2n$ and are able to Bragg reflect incoming laser radiation. In Figure A3-2, the dominant interference is between 90° scattered radiation and the forward propagating laser radiation to create iso-density fringes oriented at a 45° angle. These fringes behave as a 45° (Bragg) mirror to reflect the laser radiation at 90° . The reflected radiation and laser radiation interfere together to further amplify the isodensity fringe planes which enhances their reflectivity. The iso-density planes are phase gratings which grow exponentially to strongly reflect the incident laser radiation. The reflected radiation also grows exponentially until the incident laser radiation is depleted.

APPENDIX A-4
OPTICAL CONTROL USING STIMULATED LIGHT SCATTERING

Lauren M. Peterson
Optical Engineering, Vol. 25, No. 1
January 1986, pp. 103-107

Optical control using stimulated light scattering

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Abstract. The use of stimulated light scattering as a means for achieving optical control functions directed toward an optical computer is described. Stimulated thermal Rayleigh scattering is seen to be a preferred nonlinear optical mechanism when compared with the more familiar stimulated Raman and stimulated Brillouin scattering. It possesses the highest gain, the lowest threshold, and scattered radiation that is approximately the same frequency as the inducing radiation. Optical control functions such as optical bistable switching, optical amplification, and optical limiting or clipping are described.

Subject terms: digital optical computing; nonlinear optics; stimulated scattering; stimulated thermal Rayleigh scattering.

Optical Engineering 25(1), 103-107 (January 1986).

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5. Summary and conclusions
6. Acknowledgments
7. References

1. INTRODUCTION

Of the stimulated scattering mechanisms, stimulated Raman scattering (SRS) is probably the best known. It is used primarily as a frequency-shifting mechanism for generating new wavelengths from pulsed lasers. Stimulated Brillouin scattering (SBS) is known primarily for its undesirable characteristics of (1) inducing damage in material media, including solid-state laser rods, (2) being a limiting mechanism for SRS, and (3) limiting the amount of radiation power density that can be tolerated in fiber and integrated optical waveguides. Because the attributes of SBS have been primarily negative, work is generally directed toward eliminating its presence. Stimulated thermal Rayleigh scattering (STRS) is similar to SRS and SBS but is less well known since like SBS practical applications have yet to be demonstrated and unlike SBS it is seen only in media that are specially prepared to be adequately absorbing. In this paper, we describe the mechanism of stimulated scattering and show how STRS in particular can be put to use as an optically bistable switch, an amplifier of optical radiation, and an optical limiter.

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2. STIMULATED SCATTERING

Optical radiation passing through a material medium is weakly scattered in all directions by random fluctuations in the index of refraction. This scattering process may be viewed as a diffraction phenomenon in which the random index fluctuations are Fourier-decomposed into a three-dimensional collection of sinusoidal phase gratings representing a continuum of spatial frequencies and a continuum of orientations in space, i.e., a continuum of grating k -vectors. Random acoustic disturbances in the medium lead to phase gratings that propagate at the speed of sound, and random temperature variations in the medium lead to nonpropagating phase gratings with finite lifetimes. The former leads to spontaneous Brillouin scattering, and the latter leads to spontaneous Rayleigh scattering.

Experiments using high power ruby lasers¹⁻⁴ showed that those Fourier components that lead to backscattered radiation will beat with the forward-traveling laser radiation in the presence of a nonlinear medium to generate a density wave that precisely matches the phase grating that causes the spontaneous scattering. The magnitude of the phase grating is thereby amplified, which leads to more backscattering and so on. This process continues on a subnanosecond time scale, and if the power density of the laser is adequate and the material medium is sufficiently nonlinear, then most of the laser radiation can be redirected into the backward direction. Such intense scattering is referred to as stimulated scattering. If the amplified material wave is a propagating acoustic wave (acoustic phonon) and the radiation is Doppler-shifted, this is stimulated Brillouin scattering. If the material wave is a non-propagating thermal wave, this is stimulated thermal Rayleigh scattering. [Stimulated Raman scattering and stimulated Rayleigh wing scattering (SRWS) are analogous processes involving optical phonons and orientational fluctuations, respectively.]

2.1. Gain equations

The theory for SBS and STRS shows that electrostriction and

absorptive heating, respectively, are the mechanisms that lead to a nonlinear polarization of the material medium.⁴⁻⁶ This nonlinear polarization provides a driving term in Maxwell's wave equation and leads to coupling between the strong incident and weak scattered radiation to amplify the latter. The unsaturated amplification is found to be exponential as

$$I_s(l) = I_s(0) \exp(gI_L l) \quad (1)$$

where g is the gain coefficient for stimulated scattering, I_L is the incident laser intensity, l is the interaction length in the nonlinear medium, and $I_s(0)$ and $I_s(l)$ are the scattered radiation intensities before and after amplification, respectively.

Table I provides representative values of the gain coefficient for the different stimulated processes.⁶ Also included in the table are the magnitudes of the frequency shift $\Delta\nu$ of the scattered radiation compared to the incident laser radiation, and the lifetimes of the stimulated processes.

Although STRS is not as well known as SBS and SRS, the gain of absorption-assisted STRS is significantly greater. "Absorption-assisted" means that an absorbing dye has been added to the material medium to enhance the thermal interaction. Another feature of STRS that makes it more valuable than the other stimulated processes for optical switching and control is its small frequency shift. Whereas all of the other stimulated processes represent frequency shifts that correspond to many laser linewidths, the STRS shift is less than a single laser linewidth and for many purposes may be considered as essentially unshifted in frequency.

Herman and Gray^{4,5} have placed SBS and STRS onto a firm theoretical ground using the hydrodynamic equations modified to include the effects of electrostriction (SBS) and absorptive heating (STRS) to describe the effect of the radiation on the density of the medium. The density fluctuations couple to Maxwell's wave equation, which when solved provides an expression for the imaginary part of the nonlinear susceptibility χ_{NL} . For STRS one obtains⁵

$$\text{Im } \chi_{NL}^{\text{STRS}} = \frac{-\mu_{mol}}{M} \left(\frac{n^2 + 2}{3} \right)^2 \frac{\beta n c \alpha}{\gamma c_v} \times \frac{\Delta\omega}{(\Delta\omega)^2 + (\Gamma_L/2 + \Gamma_R/2)^2} \quad (2)$$

where μ_{mol} is the molecular polarizability, M is the molecular mass, $(n^2 + 2)^2/9$ is a local field correction, β is the thermal expansion coefficient, n is the refractive index, c is the speed of light in vacuum, α is the absorption coefficient, γ is the ratio of specific heats, c_v is the specific heat at constant volume, $\Delta\omega = \omega_L - \omega_s$ is the difference between the laser radian frequency and that of the scattered light, and Γ_L and Γ_R are the laser linewidth and spontaneous Rayleigh linewidth, respectively. The gain coefficient can be expressed as

$$g = \frac{2\pi \text{Im } \chi_{NL}^{\text{STRS}}}{\lambda_0 n^2 \epsilon_0 c} \quad (3)$$

where λ_0 is the wavelength of the radiation and ϵ_0 is the dielectric constant in vacuum.

2.2. Phase matching

In order to have gain in an optical interaction, the generated photons must be in phase with each other. Gain, and therefore

TABLE I. Typical Gain Factors g , Frequency Differences $\Delta\nu$, and Lifetimes τ of Stimulated Scattering Phenomena⁶

| | g (cm/MW) | $\Delta\nu$ (cm ⁻¹) | τ (ns) |
|---------------------|-------------|---------------------------------|-------------|
| SBS | 0.05 | 0.1 | 0.1 to 1 |
| STRS | | | |
| Transparent media | 0.0002 | 10 ⁻³ | 20 |
| Absorption-assisted | 0.2 | 10 ⁻³ | 20 |
| SRWS | 0.001 | 50 | 0.001 |
| SRS | 0.002 | 1000 | 0.01 |
| SCS* | 0.001 | 10 ⁻² | 5 |

*Stimulated concentration scattering

phase matching, are assured for stimulated scattering, provided that the laser spectral linewidth is adequately narrow in generator experiments where amplification originates from spontaneously scattered noise radiation. For amplification experiments using an injected signal, both the pump and signal radiation must be sufficiently narrow in linewidth and properly matched in frequency. A slight Stokes shift in frequency, equal to one-half the laser linewidth or less, may be required of the signal radiation in order for it to experience maximum gain.

The Fourier component of the random density fluctuation that initiates the stimulated scattering satisfies the Bragg condition⁷

$$\lambda = 2\Lambda \sin \frac{1}{2} \Theta \quad (4)$$

where Λ is the spatial period of the Fourier component and Θ is the angle through which an incident photon is scattered. The scattered photon with wave vector k_s beats with the incident radiation k_i to generate a new density wave with wave vector K satisfying the momentum matching condition

$$k_i = k_s + K \quad (5)$$

Equations (4) and (5) are equivalent since Θ is the angle between k_i and k_s and the grating vector $K = 2\pi/\Lambda$. The generated wave is identical to the initial Fourier component.

Radiation of finite linewidth produces density waves, or phase gratings, whose spatial frequencies are distributed. To assure phase matching of the scattered radiation, or equivalently to assure that the superposition of phase gratings is not washed out, the spectral linewidth of the laser radiation in wave numbers must be much less than the reciprocal of the interaction length l . This should not be a problem for most laser sources.

Phase matching of the scattered radiation is entailed in the theoretical analysis of Herman and Gray.^{4,5} It is manifest in the frequency-dependent part of Eq. (2). For an infinitesimally narrow laser ($\Gamma_L = 0$), the gain maximizes at $\Delta\omega = \Gamma_R$. This Γ_R is equal to the reciprocal of the lifetime of the thermal grating. The Γ_L that appears with Γ_R is due to the convolution of the laser profile with the ideal gain profile.

It should be stressed that the physical mechanism of the stimulated scattering interaction is essentially the same off-axis as it is on-axis for backscattering. Stimulated backscattering is routinely demonstrated primarily because spherical

lenses are commonly used that produce an axial focal volume. If optics are used that provide the longest interaction length in an off-axis direction (using an off-axis resonator⁸ or cylindrical optics), then off-axis stimulated scattering dominates.

3. OPTICAL IMPLEMENTATIONS

Stimulated scattering processes have been studied primarily with the goal of understanding the particular radiation-matter interactions involved. Although much excellent experimental work has been conducted in stimulated scattering, very little creativity has been expressed (except for SRS)⁹ in the use of multiply interacting beams, resonant cavities, or novel optical geometries in order to find useful applications for the phenomena. In the following we present optical control functions implemented with the standard on-axis optical beam geometry and also control functions implemented using cylindrical, or possibly conical, optics.

3.1. Optical bistable switching

Figure 1 shows the standard experimental configuration for generating stimulated scattered radiation. Since power densities of MW/cm² are required to see these nonlinear effects, the laser radiation is focused into the material medium. Figure 2(a) depicts the incident laser pulse intensity I as a function of time (typical times correspond to tens of nanoseconds), and the transmitted and backscattered intensities I_T and I_S , respectively. Experiments studying SRS show the existence of a distinct threshold that is characteristic of stimulated scattering, followed by strong backscattered radiation. A steady-state condition can be observed shortly after the threshold is exceeded.^{9,10}

Figure 2(b) shows that optical bistability is manifested in the stimulated interaction. As the incident radiation increases, no backscattered wave is observed until the threshold is exceeded. Beyond threshold the backscattered intensity increases with that of the incident radiation. Once the maximum I_m is reached and I falls to smaller values, I_S follows the same path until I approaches its threshold value I_{th} , at which point the downward path differs from the upward path. This region of hysteresis, or memory, is the bistable region and possesses the potential for use as an optically bistable switch.

3.2. Optical amplification

Equation (1) and the discussion above show that, in generator experiments such as depicted in Figs. 1 and 2, spontaneous noise behaves as a weak signal that undergoes orders of magnitude ($\sim e^{10}$) of amplification in generating the strong backscattered stimulated radiation. One would expect, and it has indeed been observed experimentally,^{4,6,11} that operation below threshold leads to amplification of counterpropagating signal radiation. The primary requirement for the injected signal radiation is that it be spectrally narrow and match the frequency of the strong pumping radiation. This is most easily accomplished by deriving both the pump and signal beams from the same laser source.

Amplification should also be possible in an off-axis geometry, as described in the following section.

⁹Nonlinear Raman spectroscopies such as CARS and RIKES are useful offspring of SRS that resulted from creative experiments using novel optical geometries.

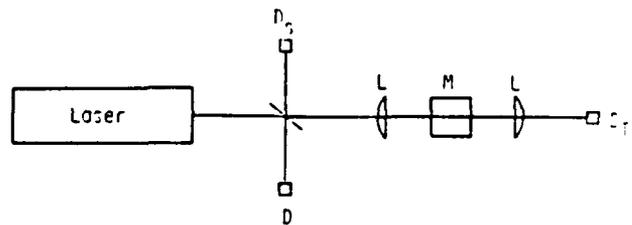


Fig. 1. Generator experiment for stimulated scattering. Detectors D , D_T , and D_S measure incident, transmitted, and stimulated radiation, respectively, for interactions in nonlinear medium M .

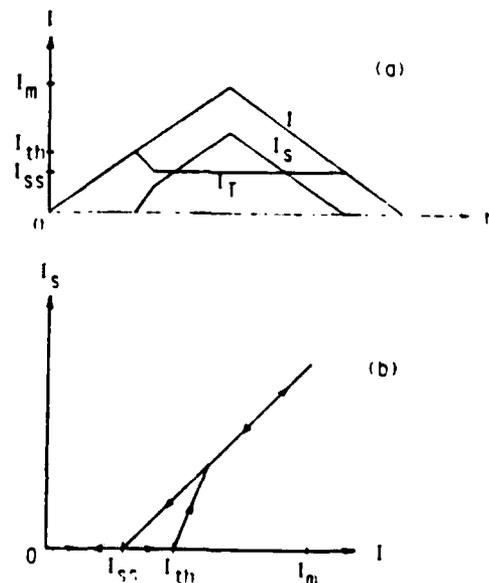


Fig. 2. (a) Temporal characteristics of stimulated scattering. I , I_T , and I_S are the incident, transmitted, and stimulated radiation, respectively. (b) Bistable characteristics of stimulated scattering.

3.3. Off-axis geometry

The various stimulated scattering mechanisms compete with each other for the laser energy, and the one with the largest gain is the one that is stimulated. Suppression of SRS in order to observe SRS is commonly known. It is less well known that a given stimulated scattering mechanism also competes with itself based upon geometrical considerations.

Most stimulated scatterings are observed on-axis because the interaction length l in Eq. (1) is greatest in the backward or forward direction such that the amplification is strongest on-axis. If a resonant cavity is placed off-axis around the interaction region such that the cavity center line establishes the direction of maximum gain, then the stimulated scattering is observed in the off-axis direction. This has been observed with SRS.⁸

The use of cylindrical, or possibly conical, optics might also provide a useful means of achieving off-axis gain and switching. Figure 3(a) depicts the line focus obtained when a beam of laser radiation is focused by a cylindrical lens. Spontaneous scattering occurs in all directions, and the highest gain is within the line focus where I is the highest. To exceed threshold for stimulated scattering, however, requires that the gain exceed the loss. The gain is highest along the direction of

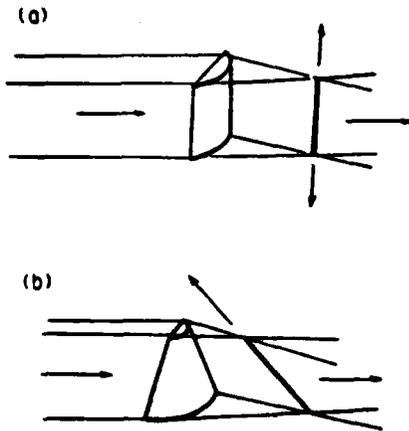


Fig. 3. Off-axis geometries for stimulated scattering using (a) cylindrical and (b) conical optics.

the line focus, and therefore threshold is first reached in a direction that is at 90° with respect to the axis of the incident laser beam.

Note that the density wave that is amplified in the interaction of Fig. 1 using spherical optics has its isodensity planes perpendicular to the directions of propagation of the optical radiation (i.e., the k -vector of the phase grating is at 0° and the k -vectors of the incident and scattered radiation are at 0° and 180° , respectively). Theory shows⁷ that gain for stimulated scattering follows a $\sin(\Theta/2)$ dependence such that backscattering dominates over forward scattering. The use of cylindrical optics utilizes (and amplifies) density waves whose isodensity planes are at 45° with respect to the laser beam axis (i.e., the grating k -vector is at 45° and the k -vectors of the incident and scattered radiation are at 0° and 90° , respectively). By symmetry, neither direction along the line focus is preferred, and bidirectional stimulated scattering is expected. A unidirectional output might be obtained by simply placing a mirror in one of the beams of scattered light in order to fold it back to the interaction region for further amplification. The use of conical optics as shown in Fig. 3(b) can also provide unidirectional off-axis output since gain is higher in the back direction than in the forward.

Switching and amplification functions can be implemented in the off-axis direction by proper choice of the magnitude of the incident or pump radiation. The off-axis geometries should exhibit those same characteristics depicted in Fig. 2. Consider the following four optical control functions:

- Self-switching.** If the pump radiation exceeds threshold, then the radiation is coupled efficiently to an off-axis direction.
- Control switching.** If the pump radiation is below threshold but above the steady-state value, then signal radiation injected along the region of highest gain switches the pump radiation from its on-axis propagation to off-axis propagation.
- Amplification.** If the pump radiation is well below threshold, then signal radiation injected along the region of gain is amplified by coupling energy from the on-axis pump beam into the off-axis direction of the signal beam.
- Optical transistor.** Operating in the amplification mode, if the pump radiation is amplitude-modulated, since the

gain is nonlinear, the stimulated scattered radiation is modulated with a greater amplitude.

3.4 Optical limiting/clipping

The last optical control function discussed is that of limiting or clipping. Figure 2 shows this most clearly. Once threshold has been exceeded, excessive radiation is converted into stimulated radiation I_s , such that the radiation remaining in the incident beam I_T is of nearly constant intensity.

4. DISCUSSION

For a control technique to be useful for optical computing applications, it must be accessible at modest optical energies. Table I shows that absorption-assisted STRS has the highest gain and therefore is the best candidate for low energy operation. The tabulated gain coefficient is 0.2 cm/MW, although a gain of 0.8 cm/MW has been observed experimentally using I_2 dye in CCl_4 liquid.^{4,11} (The measured gain coefficient for CCl_4 is in agreement with theoretical predictions.) Examination of Eq. (1) shows that since amplification is highest for high optical intensity, one might expect lowest thresholds for a tightly focused Gaussian (TEM₀₀) laser beam. A more careful examination of the focusing of a Gaussian profile beam shows, however, that the length of the focus l , which represents the interaction length, decreases as the square of the focused beam diameter due to diffraction.⁷ It can be easily shown that

$$g_{Ll} = \frac{2gP_L}{\lambda} \quad (6)$$

where P_L is the power in a TEM₀₀ laser beam of wavelength λ . Note that the exponential amplification is independent of the focused beam size and the interaction length.

Using the more conservative value for g of 0.2 cm/MW and taking λ to be 0.5 μm , significant gains should be realized for laser powers in excess of about 100 W. Actual switching should require one to two more orders of magnitude of optical power, but since the switching time for STRS is on the order of nanoseconds, the switching energy is seen to be on the order of several microjoules. These power and energy requirements are modest and may be improved upon if more efficient materials and techniques are developed.

The cylindrical geometry offers such an improvement in optical efficiency. Again assuming a TEM₀₀ laser beam and diffraction-limited optics, one can determine that

$$g_{Ll} = \frac{gP_L}{\lambda} \left[\frac{l/\lambda}{(f/D)^2} \right] \quad (7)$$

for a cylindrical lens, where f/D is the effective f -number of the cylindrical lens being used. The improvement factor of cylindrical optics compared to spherical optics is the quantity in brackets. It should be possible to make the interaction length l , here the length of the line focus, much greater than λ . If a small enough f -number can be used without deleterious effects due to diffraction, then the requirements on P_L should be considerably less than those computed above for spherical optics.

Absorption-assisted STRS is most readily attained in liquid solvents to which an absorbing dye has been added. Although inorganic and simple organic liquids (alcohols, acetone, etc.)

have been used to attain STRS, it may be possible to tailor more complex organics with desirable thermodynamic parameters such as a high thermal expansion coefficient β or a high molecular polarizability μ_{molecule} in order to increase the gain coefficient and therefore further lower the power requirements. Critical point phenomena or critical opalescence conditions may be found that drastically modify the thermodynamics such that STRS is enhanced.

5. SUMMARY AND CONCLUSIONS

Stimulated light scattering appears to be a viable method for achieving optical control functions for application to optical computing. Of the various stimulated scatterings, stimulated thermal Rayleigh scattering is clearly the best choice. It possesses the highest gain coefficient and does not suffer from large frequency shifts in its scattered radiation as do the other stimulated scatterings.

Optical control functions such as bistable switching, optical amplification, and limiting or clipping can be achieved with STRS. These can be achieved with a laser source and spherical or cylindrical focusing optics. Switching times are on the order of nanoseconds, and optical power requirements are

hundreds of watts or less. The use of cylindrical optics offers the potential for off-axis geometries and lower optical power requirements.

6. ACKNOWLEDGMENTS

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