STIMULATED RAMAN AND BRILLOUIN SCATTERING WITH PICOSECOND LIGHT PULSES

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Stimulated Raman scattering is observed to be strongly quenched under pulse excitation with pulses shorter than the molecular rotation time, because such pulses see a smaller Kerr effect and experience less self-focusing. Reduced stimulated Brillouin emission is also observed.

This Letter reports experiments showing strong suppression of stimulated Raman scattering in nitrobenzene and other liquids under excitation by picosecond light pulses. This effect is primarily due to reduction of the Kerr effect resulting in partial quenching of self-focusing for pulses shorter than the molecular reorientation time. Picosecond Raman emission is further limited by dispersion, which leads to a coherence length ~1 cm for exponential Raman gain. We also observe a large reduction in stimulated Brillouin emission.

Previous experiments[1] have demonstrated stimulated Raman conversion efficiencies of 10^{-2} to 10^{-1} in nitrobenzene, CS_2, and other liquids having high Raman-scattering cross sections. It has become clear[2-5] that the observed gains, generally in excess of the values predicted from the cross sections, can be explained by self-focusing of the pump light, arising primarily from the Kerr effect.[6]

However, suppose the light-pulse duration τ_p approaches τ, the Debye rotation time for molecules driven by an ac field. Here $\tau = 4\pi a^3\eta/3kT$, where η is the viscosity, T is the temperature, and a is the effective molecular radius.[7] When $\tau_p \gtrsim \tau$, the anisotropic molecules can rotate in response to the pulse, but when $\tau_p < \tau$, the molecules will not respond and the self-focusing threshold will increase. Values of τ are about 10^{-11}-10^{-12} sec and have been measured in liquids from observations of the frequency width of depolarized scattering near the Rayleigh line[8] and the frequency shift of stimulated Rayleigh emission.[9]

Backward stimulated Brillouin scattering will also be reduced for short pulses because the gain-length product becomes small and because damping times for hypersonic acoustic waves are much greater than a picosecond.[10]

The experiments made use of 5300-Å light pulses of picosecond duration, generated in a Nd^{+3} glass laser mode locked[11] with Eastman 9860 dye and followed by a KH_2PO_4 harmonic generator crystal. Pulse widths were measured by the two-photon fluorescence technique.[12,13] Observations of the fluorescence on mirror reflection of the pulses in 0.01M dibenzanthracene solution showed the pulse duration to be 1-2 psec. From oscilloscope displays the laser output was shown to consist of ~25 pulses separated by $\Delta t = 2L/c \sim 4 \times 10^{-9}$ sec, where L is the optical length of the laser resonator. Since $\Delta t \gg \tau$, successive light pulses can be considered to produce independent Raman scattering.

At 1.06 μ the laser output was ~1 GW which was converted to 150 MW of harmonic in a KH_2PO_4 crystal. The 5300-Å light was focused by a 30-cm focal length lens onto a 10-cm sample cell placed 25 cm from the lens. The peak laser intensity in the liquid was 10-20 GW/cm^2. Ordinarily, at this intensity efficient conversion of laser light to Raman[1] and Brillouin[14] light occurs. Table I shows relative peak intensities of Raman to laser light measured for picosecond pulse excitation in pure liquids and solutions, as recorded from phototube signals by a Tektronix 555 oscilloscope. It is evident that when $\tau_p \leq \tau$, the liquids showing most efficient stimulated Raman scattering are not those with the largest Kerr constants. With picosecond pulse excitation the data clearly show a tendency for less Raman scattering as τ becomes longer than τ_p. Nitrobenzene, previously shown to be one of the most efficient stimulated Raman emitters, was found to have the highest threshold of the liquids studied with picosecond pulse excitation; under long pulse excitation,[10] this liquid has a much lower stimulated Raman threshold than benzene and toluene. Furthermore, a nitrobenzene cell placed in front of a cell of toluene reduced the toluene Raman scattering with short pulse excitation, the opposite of the effect with long pulse excitation.[8]

To test further the correlation of conversion efficiency with molecular orientation time, stimulated CS_2 Raman scattering was measured in 1:2 solutions of CS_2 in the solvents listed

Table I. Ratios of the Raman-scattered light to the incident laser light.

Sample	Kerr constant[a] (10^{-8} esu)	Rotation time[b-e] τ (10^{-12}sec)	$(I_{\rm Raman}/I_{\rm Laser})\times10^3$	Coherence length for 1 psec pulse l_c (cm)
Carbon disulfide	32.6	1.9	3.5	1.2
Benzene	5.7	3.3	0.2	1.5
Toluene	6.5	5.3	0.02	1.7
Chlorobenzene	9.9	...	0.03	1.6
Bromobenzene	14.4	...	0.01	1.4
Nitrobenzene	26.4	50	<0.002	0.8
$\frac{1}{3}$ carbon disulfide, $\frac{2}{3}$ carbon tetrachloride	...	5	1.4	
$\frac{1}{3}$ carbon disulfide, $\frac{2}{3}$ hexane	...	...	1.1	
$\frac{1}{3}$ carbon disulfide, $\frac{2}{3}$ acetone	...	...	1.0	
$\frac{1}{3}$ carbon disulfide, $\frac{2}{3}$ bromobenzene	...	...	0.26	
$\frac{1}{3}$ carbon disulfide, $\frac{2}{3}$ nitrobenzene	...	...	<0.01	

[a] Y. R. Shen, Phys. Letters 20, 378 (1966).

[b] S. L. Shapiro and H. P. Broida, Phys. Rev. 154, 129 (1967).

[c] C. W. Cho, N. D. Foltz, D. H. Rank, and T. A. Wiggins, Phys. Rev. Letters 18, 107 (1967).

[d] I. L. Fabelinskii, Tr. Fiz. Inst., Akad. Nauk SSSR 9, 183 (1958).

[e] V. S. Starunov, E. V. Tiganov, and I. L. Fabelinskii, Zh. Eksperim. i Teor. Fiz.—Pis'ma Redakt. 4, 262 (1966) [translation: JETP Letters 4, 176 (1966)].

in Table I. The Debye relaxation formula predicts that the orientation time for CS_2 molecules should decrease with solvent viscosity through the series nitrobenzene (2 cP), bromobenzene (1.1 cP), and hexane and acetone (0.32 cP). The viscosity of pure CS_2 is 0.36 cP. Although the shear viscosity of CCl_4 is high (0.96 cP), the hindrance to CS_2 rotation is expected[15] to be small because of the spherical symmetry of the CCl_4 molecules. The measured conversion efficiencies for the solutions (Table I) show a clear inverse correlation with solvent viscosity and hence orientation times, and are uncorrelated with Kerr constants.

The backward Brillouin scattering was measured to be less than 3×10^{-4} of the incident intensity for the pure liquids in Table I. This result is to be contrasted to values from 10^{-1} to 9×10^{-1} reported with longer pulses and lower intensities.[14]

The above experiments were repeated with 35-nsec pulses from a conventional rotating-prism Q-switched ruby laser, whose output was verified by the fluorescence technique to have no significant pulse components shorter than 10 psec. At maximum intensities of ~50 MW cm^{-2} (<1% of the intensity in the psec experiments), conversion efficiency to both Raman and Brillouin light was >1% for all the liquids of Table I, nitrobenzene and bromobenzene being two of the most efficient scatterers. The solutions with highest Raman efficiencies were CS_2-nitrobenzene and CS_2-bromobenzene. These results, consistent with previous experiments in which $\tau_p \gg \tau$, show qualitative correlation of scattering efficiency with Kerr constant.

Near-field photographs of the end of the cell show that the Raman emission from picosecond light pulses occurs completely in filaments. Now at (nonself-focused) pulse intensities of 10 GW cm^{-2}, stimulated Raman gains[16] G in the range 10-100 cm^{-1} are expected[17]; however, the over-all gain is limited by dispersion, since the amplified Raman light travels faster than the pump pulse. Coincident pump and Raman pulses of duration τ_p become decoupled in a distance of the order of the coherence length $l_c \equiv \tau_p v_g^2 (2\Delta v_g)^{-1}$, where v_g is the average group velocity[13] and Δv_g is the group velocity difference of the pump and Raman pulses. For $\tau_p = 1$ psec, $l_c \sim 1$ cm for most liquids

(Table I). It follows that the over-all gain is limited to $\sim Gl_c$ or 10^1-10^2 for picosecond 10-GW pump pulses. This conclusion is supported by our failure to observe nonfilamentary stimulated Raman emission at 10-GW-cm^{-2} pump intensity over a distance of 10-30 cm.

It appears that picosecond light pulse excitation of Raman scattering may provide a valuable technique for probing molecular orientation dynamics in liquids by the use of pulse times on the scale of the molecular orientation time.

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[1] See, for example, G. Eckhardt, R. W. Hellwarth, F. J. McClung, S. E Schwarz, D. Weiner, and E. J. Woodbury, Phys. Rev. Letters 9, 455 (1962); F. J. McClung, W. G. Wagner, and D. Weiner, Phys. Rev. Letters 15, 96 (1965); D. Weiner, S. E. Schwarz, and F. J. McClung, J. Appl. Phys. 36, 2395 (1965); G. Bret, Compt. Rend. 260, 6323 (1963).

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[7] See, for example, P. Debye, Polar Molecules (Dover Publications, New York, 1929), Chap. V; N. Bloembergen, Nuclear Magnetic Relaxation (W. A. Benjamin, Inc., New York, 1961); N. Bloembergen and P. Lallemand, Phys. Rev. Letters 16, 81 (1966).

[8] See, for example, I. L. Fabelinskii, Tr. Fiz. Inst., Akad. Nauk SSSR 9, 183 (1958); V. S. Starunov, Dokl. Akad. Nauk SSSR 153, 1055 (1963) [translation: Soviet Phys.—Doklady 8, 1206 (1964)]; S. L. Shapiro and H. P. Broida, Phys. Rev. 154, 129 (1967).

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[13] P. M. Rentzepis and M. A. Duguay, to be published.

[14] E. Garmire and C. H. Townes, Appl. Phys. Letters 5, 84 (1964); R. G. Brewer and K. E. Rieckhoff, Phys. Rev. Letters 13, 334 (1964); M Maier, W. Rother, and W. Kaiser, Phys. Letters 23, 83 (1966).

[15] See, for example, W. B. Moniz, W. A. Steele, and J. A. Dixon, J. Chem. Phys. 38, 2418 (1963); R. G. Gordon, J. Chem. Phys. 43, 1307 (1965).

[16] R. W. Hellwarth, Phys. Rev. 130, 1850 (1963); Appl. Optics 2, 847 (1963).

[17] Raman gains for picosecond-pulse excitation are in fact somewhat less than calculated from the scattering cross sections, since the spectral width of the pulse is larger than the Raman linewidth. This effect cannot explain the present picosecond results, however, since for example the reduction in the gain for CS$_2$ (linewidth 1.6 cm^{-1}) would be larger than that for nitrobenzene (linewidth 8 cm^{-1}).

METHOD FOR SOLVING THE KORTEWEG-deVRIES EQUATION*

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A method for solving the initial-value problem of the Korteweg-deVries equation is presented which is applicable to initial data that approach a constant sufficiently rapidly as $|x| \to \infty$. The method can be used to predict exactly the "solitons," or solitary waves, which emerge from arbitrary initial conditions. Solutions that describe any finite number of solitons in interaction can be expressed in closed form.

For a large class of physical systems, nonlinear and dispersive processes compete while dissipation is negligible. In particular, the Korteweg-deVries (KdV) equation,

$$u_t - 6uu_x + u_{xxx} = 0 \tag{1}$$

(subscripts x and t denoting partial differentiations), has been shown to describe the asymptotic development of small- but finite-amplitude shallow-water waves,[1] hydromagnetic waves in a cold plasma,[2] ion-acoustic waves,[3] and acoustic waves in an anharmonic crystal.[4]

The quantities u, x, and t can be rescaled to produce any desired coefficients for the terms of Eq. (1). The present choice is convenient for this paper. Note that u is reversed in sign from previous work since the coefficient of