Observation of backward-stimulated Raman scattering generated by picosecond laser pulses in liquids*

R. R. Alfano and G. A. Zawadzkas
City College of New York, Physics Department, New York, New York 10031
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Stimulated Raman scattering in the backward direction is observed in organic liquids under 5300-Å picosecond excitation. The spectrum and intensity of the backward-directed light is measured and compared with the emission in the forward direction. The Stokes spectrum of the light emitted in backward direction consists of stimulated Raman light, while in the forward direction both stimulated Raman and self-phase-modulated light is detected. No stimulated Brillouin or Rayleigh light is detected in the backward direction. The experimental results are in qualitative agreement with transient Raman theory.

In recent years there has been considerable interest in transient behavior of stimulated Raman scattering. Shapiro *et al.*[1] first observed forward-stimulated Raman scattering (SRS) with picosecond laser pulses. Several investigators have demonstrated different Raman gain characteristics with transient excitation[1-5]—i.e., reduction of gain owing to quenching of self-focusing, large bandwidth of the pump, and the existence of group-velocity dispersion between the pump and Stokes waves. Extremely efficient forward Raman conversion, up to 80%, has been obtained by Colles[4] for liquids with large integrated cross sections and low dispersions. It has also been demonstrated[5] that under picosecond excitation the Stokes light emitted in the forward direction for CCl_4 has a duration that is equal to or shorter than the laser pulses. In the nanosecond regime, the properties of backward- and forward-stimulated Raman scattering in CS_2 have been measured[2], it was found that the pulse duration of the backward-stimulated Raman light is ~30 psec and the intensity ~20 times that of the laser pump wave. This effect is due to the backward-traveling Raman pulse continuously encountering undepleted oncoming pump light. Using nanosecond pulses, Loy and Shen[6] have studied and characterized self-focusing processes by investigating the dynamics of backward- and forward-stimulated Raman light in toluene.

Theoretical interest in transient SRS has been developed by many investigators.[2,7-9] The theory has most recently been extended to backward-stimulated Raman scattering[10] with picosecond pulses. The backward Raman gain[7,8,10] for short time regime in the strong-coupling limit ($\Gamma t_p < \ln G$) is given by

$$\ln G = (bI_0 V_0 \Gamma)^{1/2} t_p , \tag{1}$$

and in the weak-coupling limit ($\Gamma t_p > \ln G$) by

$$\ln G = \tfrac{1}{2} bI_0 V_0 t_p , \tag{2}$$

where b is the steady-state gain (cm/MW) (usually denoted by g [2,4]), Γ^{-1} is the dephasing time of the molecular vibrations, and I_0, V_0, and t_p are the intensity (MW/cm^2), velocity, and pulse duration of the laser pulse, respectively. Effectively, the backward gain is over the length of the pump picosecond pulse ~1 mm; thereafter the pump and backward Raman pulse become decoupled. The calculated interaction length in the forward direction for ethanol and methanol is ≈12 cm for 4.5-psec pump pulses. Recently, Kelley and Gustafson[10] proposed backward-stimulated Raman and Rayleigh scattering generated with ultrashort laser pulses as the possible loss mechanism required to explain the observed limiting diameter sizes of filaments.

This paper reports the first measurement of backward-stimulated Raman scattering from molecular vibrations in methanol, ethanol, and benzene with picosecond laser pulses. Under the high power the Stokes spectrum of the light emitted in the backward direction consists of stimulated Raman light, while in the forward direction both stimulated Raman and self-phase modulated light is detected. This scattering is observed in a time regime where the laser's pulse duration t_p is on the order of the dephasing time Γ^{-1} of the molecular vibration ($\Gamma t_p < 10$). The effective backward gain occurs over the pump pulse duration. New experimental characteristics of the transient stimulated Raman process and filaments are presented. The observed backward-stimulated light is interpreted in terms of the most recent transient-stimulated Raman gain theory.[10] The weak intensity of the observed backward-

stimulated Raman signals points to some other mechanism to explain the limiting diameter size of a filament.

The experimental arrangement for observing the backward- and forward-stimulated Raman scattering is shown in Fig. 1. A Nd-glass mode-locked laser is used to generate picosecond light pulses which are then converted in a potassium dihydrogen phosphate crystal to second-harmonic pulses at 5300 Å. The pulse was measured to be 4.5 psec by the two-photon fluorescence technique (TPF). The second-harmonic pulses of power $\sim 3 \times 10^8$ W are reduced in size to a collimated ~1-mm-diam beam in the samples by an inverted Galilean telescope. The samples are placed in a rectangular cell with optical path length 15 and 20 cm wide, oriented at a large angle of 20° to the incident propagation direction to prevent spurious reflections from the cell walls from traveling backwards along the incoming beam direction. The backward-directed light is reflected off a wedge beam, splitting it into a spectrometer. The forward-directed light is delayed relative to the backward-directed light by approximately the cavity length and directed into the same spectrometer at a slightly different slit height. This allows observation of the forward- and backward-emitted light simultaneously by either photographing the spectrum or detecting the interleaved pulse trains with a TRG 105 B photodiode. The spectra are recorded on Polaroid-type 57 film and the photodiode signals are displayed on Tektronix 7904 scope. A Corning 3-67 or 3-68 filter and neutral-density filter are used to reduce the intensity of the forward-directed light to a level comparable to that of the backward-traveling SRS light.

Spectra showing both the forward (top) and backward (bottom) emission from ethanol (a) and methanol (b) are shown in Fig. 2. The following salient features are evident in the spectra: the spectrum of the forward light (top) has both stimulated Raman emission lines and a broad band emission spanning the Stokes side—identified previously as self-phase-modulated light.[11] The exciting light at 5300 Å traveling in the forward direction is reduced by Corning and neutral-density (ND) filters. The spectra of the backward-directed light exhibits only SRS light with no light centered about 5300 Å. In ethanol the Stokes line at 2928 cm^{-1} and in methanol two Stokes lines[4,5] at 2834 and 2944 cm^{-1} are observed. The lack of light at 5300 Å in the backward direction indicates that no detectable amount of stimulated Brillouin light or Rayleigh light is produced in that direction. Figure 3 shows interleaved forward and backward Stokes SRS light from ethanol. The intensity ratio of the forward-to-backward SRS is $\sim 10^4$ as measured electronically and spectroscopically with neutral density filters. Forward SRS is observed to appear before the backward SRS. In benzene the Stoke line at 992 cm^{-1} is observed in the backward direction, while the Stokes line at 3064 cm^{-1} and first and second Stokes lines of 992 cm^{-1} vibration are observed in the forward direction. Self-focusing occurs with approximately 30–100 filaments formed with diameters ranging in size from 20 to 80 μm. Precise measurement of the forward gain is not possible with the filamentary structure. Measurement of the pulse duration of the backward SRS was not possible by TPF technique because of insufficient intensity of the pulse. An estimation of <800 psec for an upper limit of the pulse duration was made by the photodiode-oscilloscope combination.

The transient backward Raman gain for 2928 cm^{-1} line in ethanol is estimated to be $\ln G \sim 19.3$ for b

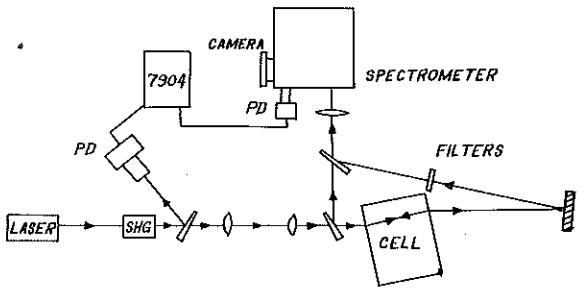

FIG. 1. Experimental arrangement for observation of forward- and backward-stimulated Raman scattering.

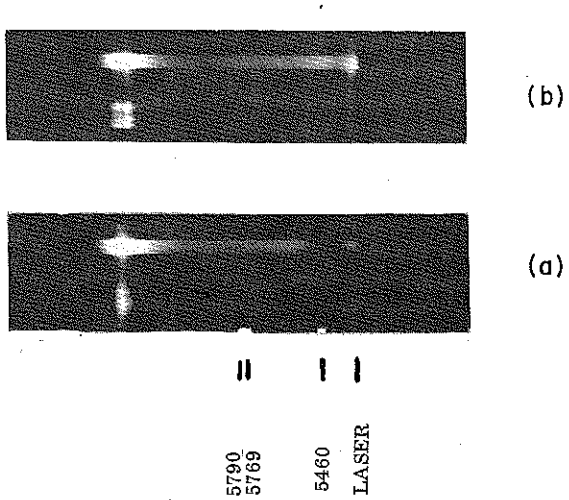

FIG. 2. Stokes spectrum emitted from ethanol (a) and methanol (b) in the forward (top) and backward (bottom) directions upon the passage of intense 5300-Å psec pulses. Filters used to take forward-direction photograph: (a) 1-(3-67) and ND = 2.6; (b) 2-(3-68) and ND = 2.6.

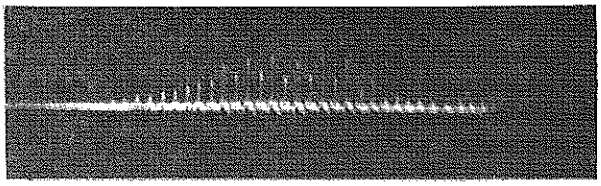

FIG. 3. Interleaved Stokes SRS pulse trains traveling in the forward and backward directions. The forward-SRS light is reduced by neutral density and Corning filters by 2×10^{-4} and is the more intense train in this photograph.

$=5.1\times10^{-3}$ cm, effective intensity in the filament $I_0\sim10^5$ MW/cm^2, $V_0\sim2.25\times10^{10}$ cm/sec, $\Gamma=1.64\times10^{12}$ sec^{-1}, $t_p=4.5\times10^{-12}$ sec, $\Gamma t_p=7.38$. This calculation shows that backward Raman gain is clearly large enough at mode-locked laser power in a filament to observe stimulated Raman scattering in the backward direction. From the observed ratio of forward-to-backward SRS intensity ~10^4, an effective filament length of ~2.2 mm is calculated[12] from ratio of forward-to-backward SRS intensity given by

$$I_F/I_B\cong\exp[(bI_0\Gamma t_p)^{1/2}(\sqrt{l_F}-\sqrt{l_p})]\,, \qquad (3)$$

where l_F is the effective length of filament and l_p is the length of the pulse for $\Gamma t_p<\ln G$.

The observation of the backward SRS with picosecond pulses is quite important since it may be one of the loss mechanisms required to explain the experimentally observed limiting diameter size [5,11,13-15] in the focal region. With the pulse duration comparable to the nonlinear Kerr-index response time, the pulse becomes deformed through the dynamics of self-focusing into a horn-shaped region,[10,16-18] with the leading edge being focussed least and the degree of focusing increasing as the peak of the pulse is approached. Unless a loss mechanism which depletes the intensity in the tail of the pulse is operative, the pulse will undergo a catastrophic focus.[10,19] A recently proposed model[10] suggests that backward-stimulated processes may provide this loss mechanism. Although we report the observations of backward SRS, it would appear that its extremely low intensity, $\sim2\times10^{-5}\,I_L$, would not provide the required loss. Some other mechanism must be responsible for the loss—possibly a multiphoton process such as two- or three-photon absorption or four-photon parametric emission.[11,20]

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[12]This calculated length is shorter than that observed for nanosecond pulses (Ref. 13) of few cm. The termination of a filament may be attributed partly to SRS-induced heating of the medium multiphoton absorption (Ref. 14). In addition, the *backward* SRS can limit the dimensions of a filament by depleting the trailing edge (Ref. 16) of the pump pulse, and consequently quenching the self-focusing action.
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