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INTENSE LIGHT BURSTS IN THE STIMULATED RAMAN EFFECT

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This Letter reports the observation in backward stimulated Raman scattering of short pulses of Stokes radiation having a peak power one order of magnitude higher than the incident laser pump power. Direct observation shows the pulses to have a duration $\sim 3\times10^{-11}$ sec ($\sim$1 cm). This effect has been observed most strongly in carbon disulfide. Previous studies[1-4] of backward stimulated Raman scattering have reported the time-average backward Stokes power equal to or less than the forward power, both being a small fraction of the incident laser power. The intense pulses observed in this work are shown to be analogous to the sharpened wave fronts which arise in propagation through an amplifying medium having an inverted population.[5-10] In forward-traveling wave-stimulated Raman gain, pulse generation is prevented by pump saturation. A given traveling volume element of Stokes wave has access only to pump energy stored in approximately the same volume, since both waves travel at about the same velocity. In backward wave gain, on the other hand, the leading edge of the Stokes wave can extract pump energy stored throughout the amplifying region, attaining a transient peak power far in excess of the pump power. Because of the large available gain and suitable initiation conditions, the stimulated Raman effect may provide a generally useful source of highly intense picosecond light pulses.

Our experimental arrangement is shown in Fig. 1. The pump source is a Q-switched ruby laser having a nearly diffraction-limited angu-

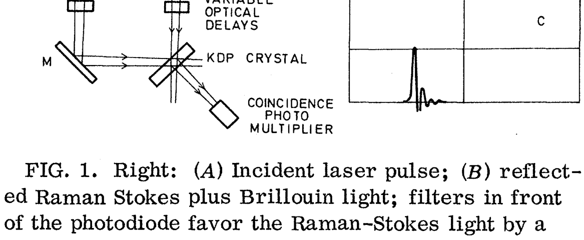

FIG. 1. Right: (A) Incident laser pulse; (B) reflected Raman Stokes plus Brillouin light; filters in front of the photodiode favor the Raman-Stokes light by a factor of 3; (C) reflected Raman-Stokes light only. (20 nsec per division.) Left: Schematic of the experimental arrangement to measure the length of the Raman spikes.

lar divergence of 5×10^{-4} rad, peak power of 1.4 MW, and pulse duration of 12 nsec. The laser beam passes through a 3× inverted telescope into a 30-cm cell of carbon disulfide. The 4.3-m separation of the laser and the cell was adequate to eliminate multiple pulses of amplified Brillouin light. The incident, reflected, and transmitted light signals were measured with photodiodes and Tektronix 519 oscilloscopes, the over-all detection systems having a rise time of ~0.5 nsec. Figure 1 shows oscilloscope traces of the incident laser light (A), the reflected Stokes plus Brillouin light (B), and the reflected Stokes light only (C).

The backward (reflected) Raman-Stokes component appears reproducibly as a single pulse occurring several nanoseconds after the beginning of the laser pulse, with a width <1 nsec and a sufficiently high energy to produce ringing of the detection system. The apparent peak Stokes power of 5×10^{5} W is ~2 times the instantaneous laser power and represents a lower limit to the true peak value. The forward Stokes component consists typically of 5-10 irregular spikes of duration 1 nsec or less, similar to those previously reported.[11,12] The apparent peak power in the forward direction is of the order of 1% of the peak laser power. Near-field photographs of the exit cell surface showed that the forward Stokes light occurs primarily in filaments, in agreement with Refs. 11 and 12 and Garmire, Chiao, and Townes.[13] On the other hand, almost all of the intense reflected Stokes pulse is emitted from a roughly uniform broad area of diameter 0.7 mm coinciding with the area illuminated by the incident laser beam. The frequency shift of the backward emission is 6.5×10^{2} cm^{-1}, in agreement with previous observations.

The backward Stokes pulse is immediately followed by (or is coincident with) the abrupt onset of stimulated Brillouin scattering, which reaches a peak value of 92% of the incident laser power, and continues throughout most of the laser pulse.[14]

Direct measurements of the Stokes pulse duration were made with the intensity autocorrelation technique[15] shown in Fig. 1. The backward Stokes pulse $I(t)$ is divided at the beam splitter into two components (1, 2) which, after undergoing a variable differential optical delay time τ, are subsequently recombined in a crystal of potassium dihydrogen phosphate (KDP) oriented to allow phase-unmatched second harmonic generation. In our experiments the time delay τ was provided by suitable combinations of glass plates and liquid cells. The coincidence photomultiplier detects the harmonic-beam component having propagation vector $\vec{k}_1+\vec{k}_2$ and therefore provides an integrated signal $J(\tau)$ proportional to $\int I(t)I(t+\tau)dt$. The pulse duration was taken to be the value of τ for which $J(\tau)/J(0)=0.5$. From several sets of measurements the average Stokes pulse duration was found to be $\tau_S=3\times10^{-11}$ sec. The time resolution of the experiment, determined by the beam and crystal geometry, was 1×10^{-11} sec. From an independent measurement of the pulse energy, 2×10^{-4} J, the peak pulse power is estimated to be 3 MW, or nine times the instantaneous laser power. The observed pulse energy represents about 1/5 the maximum available energy stored in the total cell length.

In an independent experiment, τ_S was estimated from relative energy measurements of second harmonic light generated from the Stokes and Brillouin pulse ($\tau_B=1\times10^{-8}$ sec).[16] The energy ratio of the fundamental pulses, which had equal geometrical cross sections, was $E_1(\text{Stokes})/E_1(\text{Brillouin})=0.012$; for the harmonic pulses, $E_2(\text{Stokes})/E_2(\text{Brillouin})=0.03$. If one assumes similar pulse shapes, these data lead to an estimate of $\tau_S=5\times10^{-11}$ sec in reasonable agreement with the first experiment.

The initial backward-wave Stokes amplification is described by the rate equations

$$\frac{\partial n}{\partial t}+c\frac{\partial n}{\partial x}=\sigma c\Delta n, \tag{1}$$

$$\frac{\partial \Delta}{\partial t}-c\frac{\partial \Delta}{\partial x}=-\sigma c\Delta n, \tag{2}$$

where n and Δ represent the Stokes and laser photon densities, respectively, propagating with speed c in the $+x$ and $-x$ directions, and σ is the stimulated emission cross section. The general solution[17] of Eqs. (1) and (2) shows the growth of a sharp pulse near the leading edge of the Stokes wave. Consider the special case of uniform laser photon density Δ_0, where the unsaturated gain seen by the leading edge of the Stokes pulse is $\sigma\Delta_0\equiv G$ cm^{-1}. From a model in which the initial Stokes wave has linearly increasing intensity, the calculated pulse width at half-maximum intensity (in CS_2) is $c\tau_S \sim 6.9/RG$ cm, where $R\Delta_0$ is the peak Stokes photon density, and $R\gg1$. The calculated pulse energy is $\sim(5.2/\sigma)\ln(2.9R)$ photons cm^{-2}. From the values $\sigma = 5.2\times10^{-17}$ cm^2,[18] beam diameter 0.7 mm, $\Delta_0 = 1.5\times10^{16}$ cm^{-3} at the time of the pulse, and $c\tau_S = 5.5\times10^{-1}$ cm (in CS_2), one calculates from the ratio (pulse energy)/(pulse width) the power gain $R = 9$ quoted above, and from the product (gain)$\times$(pulse width) the value $R = 16$. A third estimate, $R = 10$, follows from the harmonic generation experiments. The three independent estimates are consistent to within the experimental accuracy. The calculated pulse energy is 3×10^{-4} J, in agreement with the observed energy.

A probable mechanism for the initiation of the pulse is the abrupt onset of backward stimulated Stokes emission near the exit cell surface, accompanying the occurrence of laser self-focusing in that region. In measurements of the dependence on cell length of the laser threshold power for pulse formation, it was, in fact, found that a plot of (laser threshold power)$^{1/2}$ versus (cell length)$^{-1}$ gave a straight line characteristic of the self-focusing effect,[19] a result supporting the above suggestion. A mechanism contributing to the observed quenching of subsequent backward pulses is the build-up of intense stimulated Brillouin scattering following the pulse. Two-photon absorption does not appear to be important in the present experiments.[14]

The rate equations are inadequate to describe pulses of length less than $(2\pi\Delta\bar{\nu})^{-1} = 5\times10^{-2}$ cm, where $\Delta\bar{\nu} = 3$ cm^{-1} is the full width of the spontaneous Raman line. Such pulses may occur with longer cells and improved geometry. The analyses of Wittke and Warter,[20] which take explicit account of the dynamic macroscopic polarization, can be generalized to describe stimulated Raman pulses. A preliminary conclusion is that pulses of length significantly narrower than $(2\pi\Delta\bar{\nu})^{-1}$ can be generated with a limiting energy of the order of Δ_0/α photons cm^{-2}, where α is the residual linear absorption per cm.

Pulses of the type described above may play a role rather generally in high-gain stimulated Raman and Brillouin experiments, although the repetitive occurrence of such short pulses may not be readily apparent with conventional detectors. Such picosecond light pulses may be useful in nonlinear optical experiments and other applications. Since the occurrence of a backward pulse momentarily exhausts the laser pump light, this effect may contribute to the instability of self-trapped filaments and to the irregular nanosecond spiking observed in stimulated Raman scattering.[11,12]

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