

# Stimulated Brillouin Scattering by a Multi-Mode Pump with a Large Number of Longitudinal Modes

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We report that a multi-mode pulse with a large number of longitudinal modes shows enhanced stimulated Brillouin scattering (SBS) due to self-focusing, so the SBS reflectivity for the multi-mode pump can be higher than that for a single-mode pump case near the SBS threshold, even in the transient regime. We think that the self-focusing is generated by the high-intensity spikes of the multi-mode pulse created by mode beating, which are absent in a single-mode pulse. Optical breakdown and nonlinear effects generated by the intensity spikes, however, suppress SBS at high pump energies.

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## I. INTRODUCTION

Recently, stimulated Brillouin scattering (SBS) has been widely used in laser systems because SBS offers a powerful technique for producing a phase conjugate wave and pulse compression [1–6]. In fact, the SBS reflectivity is nearly equal to that of an ordinary mirror in the narrow band case wherein the pump bandwidth  $\Delta v_p$  is smaller than the Brillouin line-width  $\Gamma$ , which corresponds to a steady-state SBS [7]. However, from a practical point of view, many applications of a SBS phase conjugate mirror (PCM) would necessarily involve a broadband-pumped SBS, which corresponds to a transient state SBS ( $\Delta v_p > \Gamma$ ), because laser systems involving the use of an SBS-PCM have a broadband spectrum, in general, in order to obtain high output power and short pulse widths [4, 5]. Thus, it is particularly important to investigate broadband-pumped SBS. Several theoretical and experimental investigations have been reported on the SBS reflectivity using a broadband pump [8–14].

For a broadband pump, the SBS reflectivity depends on the relationship between four parameters: the coherence length  $l_c$ , the characteristic interaction length  $z_0$  which is usually equal to the Rayleigh range, the mode spacing  $\Omega_m$ , and the Brillouin line-width  $\Gamma$  [8–14]. We investigated the reflectivity of SBS-PCM when the coherence length is longer than the interaction length

( $l_c > z_0$ ). In that regime, the SBS gain for the broadband pump is as high as that for the narrow band pump [8–10]; hence, the SBS-PCM is most likely to apply to a laser system satisfying the condition  $l_c > z_0$ . The SBS gain was reported to be the same as that for a single longitudinal mode pump and to be independent of the mode structure if the pump laser mode spacing exceeds the Brillouin line-width ( $\Omega_m > \Gamma$ ) [8]. Besides, even if  $\Omega_m < \Gamma$ , off-resonant acoustic waves, which are generated by the beating between the pump laser mode and another Stokes mode, play an important role in enhancing the gain and the reflectivity [11, 12].

In all previous works, however, the influence of the multi-mode pump, or in other words a broadband pump, was considered only for two or several longitudinal modes and a low pump energy near the SBS threshold. We have found that the SBS reflectivity is significantly affected by self-focusing in the case of a multi-mode pump with a large number of longitudinal modes because the multi-mode pulse has temporal intensity spikes, arising due to mode beating, which have enough power to induce nonlinear effects, such as self-focusing and optical breakdown, even near the SBS threshold. In this paper, we investigate the characteristics of the SBS reflectivity by using a multi-mode pump with a large number of longitudinal modes over a wide range of energy, in contrast to the single-mode pump case. We used four kinds of liquids with different nonlinear refractive indices  $n_2$  and Brillouin line-widths  $\Gamma$ .

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## II. EXPERIMENTS

The experimental setup for measuring the reflectivity of the SBS-PCMs is shown in Fig. 1. The pump laser used was a Q-switched Nd<sup>3+</sup>:YAG laser (GCR-150-10, Spectra Physics) with a single-longitudinal mode injection seeder. This pump laser can be operated both in single mode and multi-mode. The laser line-width in the single-mode and the multi-mode cases were found to be  $\sim 0.003 \text{ cm}^{-1}$  (0.09 GHz) and  $\sim 0.33 \text{ cm}^{-1}$  (10 GHz), respectively. Thus, the line-width of the multi-mode case was much larger than the Brillouin line-width of the liquids used in this experiment, which are listed in Table 1 [6,7,15,16], implying that the multi-mode pump corresponded to the broadband pump and the high transient regime. The mode spacing  $\Omega_m$  was  $\sim 0.0061 \text{ cm}^{-1}$  (183 MHz); hence, the number of modes of the multi-mode pump beam was as many as about 55. The pulse repetition rate was fixed at 10 Hz, and the radius of the output beam was 4 mm. The focal length of the lens used was 15 cm. This corresponded to a Rayleigh range  $z_0$  of  $\sim 0.62$  mm. The coherence length  $l_c$  was found to be  $\sim 3.4$  cm, which was measured directly using Michelson interferometer. Thus, the coherence length satisfied the condition  $l_c \gg z_0$ . The pulse width in both cases, single-mode and multi-mode cases, was 6  $\sim$  8 ns (FWHM). The spatial beam profiles in both cases were also almost the same and nearly Gaussian. The pump energy was varied from zero to 400 mJ by means of an attenuator, which consisted of a thin film polarizer (Pol) and a half-wave plate ( $\lambda/2$ ). The output of the oscillator was reflected by using a polarization beam splitter (PBS) and was focused into the SBS-PCM. Because the beam reflected by the SBS-PCM had P-polarization after having passed through a quarter-wave plate ( $\lambda/4$ ), it passed through the PBS.

The pulse shapes of the pump and the reflected beams were measured simultaneously by using a fast photodiode. A neutral density filter (ND) was used to attenuate

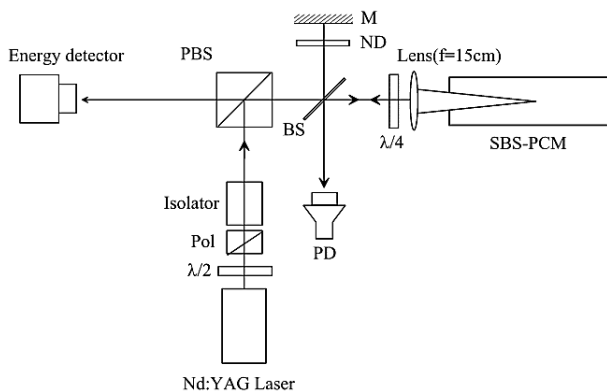


Fig. 1. Experimental setup for the measurement of the SBS reflectivity:  $\lambda/2$ , half-wave plate; Pol, polarizer; M, mirror; ND, neutral density filter;  $\lambda/4$ , quarter-wave plate; PBS, polarizing beam splitter; BS, beam splitter; PD, photodiode.

the energy of the pump pulse. The average fluctuation of the laser energy was less than 1 % for both cases. A calorimeter (Astral<sup>TM</sup> AC2501, Scientech) was used to measure both the pump energy  $E_p$  and the reflected energy  $E_R$ , which were averaged over  $\sim 300$  serial pulses. The SBS materials used in this experiment were Fluorinert FC-75, carbon tetrachloride ( $\text{CCl}_4$ ), acetone, and carbon disulfide ( $\text{CS}_2$ ). The SBS properties and the nonlinear refractive index  $n_2$  of each liquid are shown in Table 1. They have different nonlinear refractive indices  $n_2$  ranging from 0.34 to  $122 \times 10^{-22} \text{ m}^2/\text{V}^2$ . Besides, each liquid has a different Brillouin line-width ranging from 50 to 528 MHz. The breakdown threshold  $E_b$  listed in Table 1 was measured when a bright spark appeared inside the SBS cell. Each liquid was purified with a syringe filter having a 0.2- $\mu\text{m}$  pore size to raise the breakdown threshold energy  $E_b$ . The length of the SBS cell was 30 cm. The cells were composed of a glass tube and windows that were AR coated on one side only.

## III. RESULTS AND DISCUSSION

Figures 2 and 3 show the SBS reflectivity for both the single- and the multi-mode cases in  $\text{CCl}_4$  and Fluorinert FC-75 as a function of the pump energy. For both liquids, peak values exceeding 90 % can be obtained with the single-mode pump. The SBS reflectivity shows a typical nonlinear variation with the pump energy as in the case of steady-state SBS. For the multi-mode pump, the SBS reflectivity is different for each of the liquids. The SBS gain for the multi-mode pump seems to be as high as that for the single-mode pump in both the liquids, considering that the SBS threshold for both the pump modes is approximately the same, regardless of the liquid. However, in  $\text{CCl}_4$ , the peak reflectivity is 30 % at most. FC-75 provides a peak reflectivity over 65 %, but

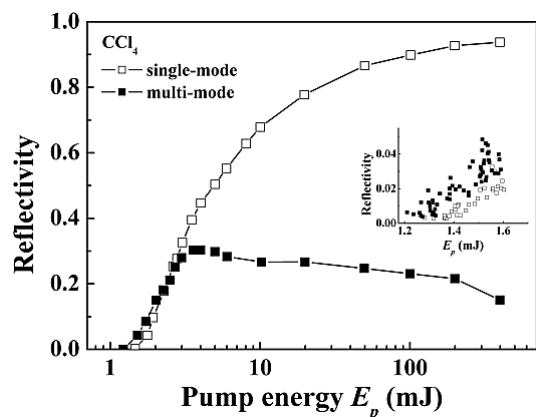


Fig. 2. SBS reflectivity vs. pump energy for  $\text{CCl}_4$  as the active medium and for laser radiation in the single-mode and the multi-mode cases. The inset in the figure represents the SBS reflectivity near the SBS threshold.

Table 1. Properties of the liquids used for experiments at a wavelength of 1  $\mu\text{m}$ .

Liquid	$\Gamma$ (MHz)	$g_B$ (cm/GW)	$n_2$ ( $10^{-22}$ m <sup>2</sup> /V <sup>2</sup> )	$P_c$ (MW)	$E_b$ (mJ)
Fluorinert FC-75	350	4.5 ~ 5	0.34	7.0	6
Carbon tetrachloride (CCl <sub>4</sub> )	528	3.8	5.9	0.4	1.7
Acetone	119	15.8	8.6	0.28	1.5
Carbon disulfide (CS <sub>2</sub> )	50	68	122	0.020	< 0.3

$\Gamma$ , Brillouin line-width;  $g_B$ , steady state SBS gain;  $n_2$ , nonlinear refractive index;  $P_c$ , critical power for self-focusing (calculated);  $E_b$ , breakdown threshold energy (measured).

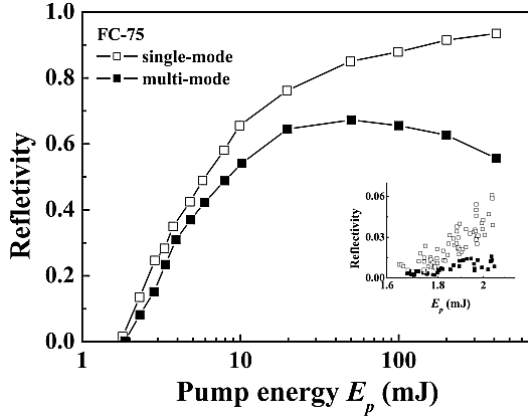


Fig. 3. SBS reflectivity *vs.* pump energy for FC-75 as the active medium and for laser radiation in the single-mode and the multi-mode cases. The inset in the figure represents the SBS reflectivity near the SBS threshold.

the reflectivity decreases as the pump energy increases.

The insets in Fig. 2 and Fig. 3 show the SBS reflectivities near the SBS threshold, which were measured for a single pulse as a function of the pump energy by using two pyroelectric detectors (70271 and 70273, Oriel). The SBS reflectivity in CCl<sub>4</sub> is slightly higher for the multi-mode pump than it is for the single-mode pump near the SBS threshold although, in general, the single-mode pump has a higher SBS gain. However, for FC-75, the behavior is exactly the opposite. Note that both CCl<sub>4</sub> and FC-75 have very similar SBS properties, such as the SBS gain and Brillouin line-width (see Table 1), which results in similar reflectivities for both liquids for the single-mode pump case. Several factors appear to contribute to the SBS for the multi-mode pump case. We interpret the SBS reflectivity for the multi-mode pump in terms of the temporal intensity spikes of the multi-mode pulse, which are absent in the single-mode pulse, because the intensity spikes created by beating between a large number of longitudinal modes have enough power to induce optical breakdown and nonlinear effects, such as self-focusing and stimulated Raman scattering (SRS). In our experiments, the multi-mode pulse is composed of 55 longitudinal modes, implying that the peak intensity of the multi-mode pump can be 55 times higher than that of the single-mode pump if perfect mode-locking is

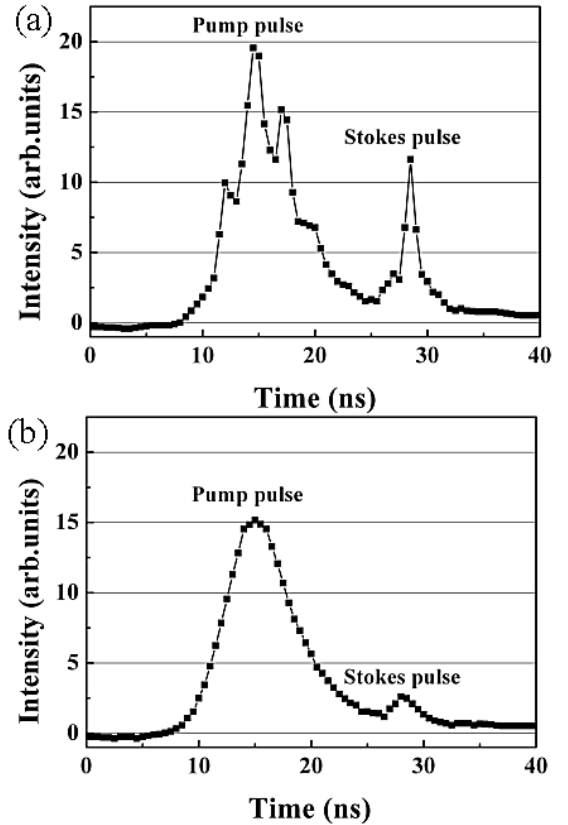


Fig. 4. Pump and reflected pulse shapes in the (a) multi-mode and the (b) single-mode cases at  $E_p \sim 1.5$  mJ in CCl<sub>4</sub>.

achieved.

First of all, the self-focusing due to the intensity spikes is likely to lead to anomalously high reflectivity of the multi-mode pump near the SBS threshold in CCl<sub>4</sub>. The critical power of the self-focusing is given by

$$P_c = \frac{\pi\epsilon_0 c^3}{n_2 \omega^2}, \quad (1)$$

where  $\epsilon_0$  and  $c$  are the permittivity of vacuum and the speed of light, respectively [17]. Using Eq. (1), the critical power  $P_c$  for CCl<sub>4</sub> and FC-75 are 0.4 and 7.0 MW, respectively; the nonlinear refractive index is listed in Table 1. Compared to the SBS threshold ( $\sim 5\%$  energy reflection) of the single-mode pump case for CCl<sub>4</sub>, which is approximately 1.8 mJ (0.26 MW), the critical power

of 0.4 MW is slightly large. However, we believe that it is possible for the multi-mode pulse to induce temporal small-scale self-focusing in  $\text{CCl}_4$  below the SBS threshold because the intensity spikes can have high peak powers exceeding the critical power  $P_c$ . Figure 4 represents the temporal pulse shapes of the pump and the Stokes pulse with different energy scales; the pump beam with  $E_p \sim 1.5$  mJ was focused into a  $\text{CCl}_4$  cell. This was measured using a fast photodiode (818-BB-30, Newport) connected to a Hewlett Packard 54542C digital oscilloscope. As we expected, the multi-mode pulse had large intensity spikes, which did not show up in the single-mode pulse. In addition, self-focusing is well known to depend on the power and not the energy of the light. Thus, the multi-mode pulse can induce self-focusing in  $\text{CCl}_4$  due to the high intensity spikes even at powers lower than the SBS threshold. Self-focusing leads to an increase in the intensity of the pump beam in the focal region and, hence, can reduce the SBS threshold energy, assuming that the steady-state SBS threshold relationship  $I_{th}g_B l = 25 - 30$  holds to a good approximation, where  $I_{th}$ ,  $g_B$ ,  $l$  are the SBS threshold intensity, the SBS gain, and the interaction length, respectively [18]. Consequently, self-focusing results in a lower SBS threshold and slightly higher reflectivity near the SBS threshold in  $\text{CCl}_4$ , as shown in Fig. 2. It is noted that backward stimulated Raman scattering (SRS) was not observed in this low-energy region up to 10 mJ although the SRS process has a very short response time of  $10^{-11}$  second, implying that it can respond better than the SBS process to the intensity spikes of the multimode pulse [19].

On the other hand, the SBS reflectivity in FC-75 is not affected by the self-focusing near the SBS threshold because the critical power for FC-75 is approximately 18 times larger than for  $\text{CCl}_4$ . Consequently, the SBS reflectivity for the multimode pump is lower than that for the single mode pump near the SBS threshold. The self-focusing seems to be deleterious for SBS since it can enhance optical breakdown. It can be shown from the experimental results that the optical breakdown in  $\text{CCl}_4$  starts at  $E_p \sim 1.7$  mJ while it begins at  $\sim 6$  mJ in FC-75, as listed in Table 1. We have observed that as the pump energy increases, the breakdown appears around the focal spot near the breakdown threshold, becomes severe, and results in a filament consisting of bright sparks. This breakdown disturbs the creation of the acoustic phonon. Therefore, for the multi-mode pump, FC-75, which has a relatively small  $n_2$ , provides a higher SBS reflectivity than  $\text{CCl}_4$  at high pump energies. On the other hand, for the single-mode pump, the reflectivities of both liquids are almost the same because no optical breakdown occurs up to a pump energy of 400 mJ.

In addition to the breakdown due to self-focusing, an intensity spike can easily generate optical breakdown by itself because it has a very steep rising edge. It is well known that for efficient SBS to occur, temporal fluctuations in the pump must be slow compared to the acoustic phonon lifetime. If the temporal fluctuations are

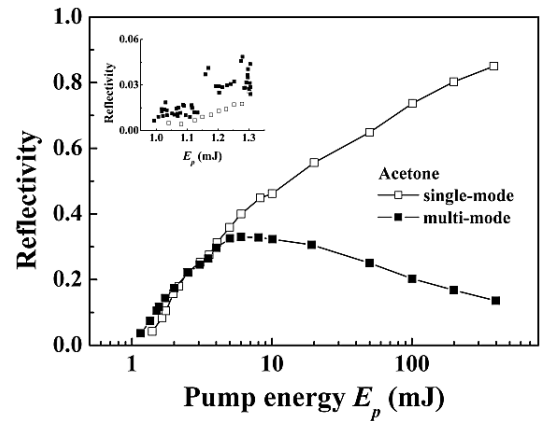


Fig. 5. SBS reflectivity *vs.* pump energy for acetone as the active medium and for laser radiation in the single-mode and the multi-mode cases. The inset in the figure represents the SBS reflectivity near the SBS threshold.

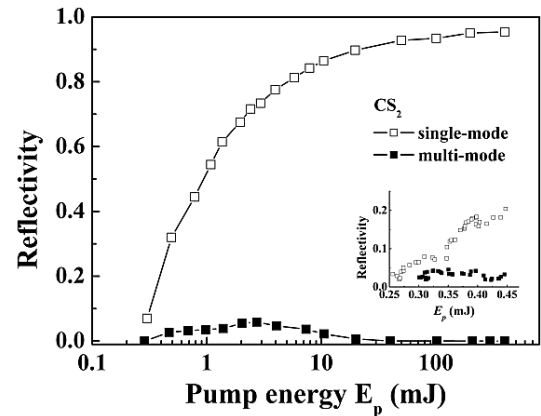


Fig. 6. SBS reflectivity *vs.* pump energy for  $\text{CS}_2$  as the active medium and for laser radiation in the single-mode and the multi-mode cases. The inset in the figure represents the SBS reflectivity near the SBS threshold.

fast relative to the acoustic phonon lifetime, the acoustic waves do not have time to build up. The intensity spikes with the steep rising edges and with energies exceeding the breakdown threshold can, therefore, reach the focal area without losing their energy by backward reflection. Hence, they can generate optical breakdown and reduce the SBS reflectivity even at low energy. For the single-mode case, the region of SBS reflection moves rapidly, along with the SBS pulse, opposite to the pump pulse, and the pump pulse is reflected well from a region before the focal area wherein the optical intensity is too small to induce optical breakdown [20]. Therefore, optical breakdown is not generated for the single-mode pump even with a strong pump energy. In addition, the SBS reflectivity for the multi-mode pump is likely to decrease due to the SRS at the high pump energy because the SRS process with a short response time can respond to the intensity spikes.

Figure 5 shows the SBS reflectivity for acetone. It

shows that the multi-mode pump provides higher reflectivity than the single-mode pump near the SBS threshold, which is very similar to the results in the case of  $\text{CCl}_4$ . The backward SRS was not observed in this low-energy region. From Table 1, acetone has approximately the same nonlinear refractive index as  $\text{CCl}_4$ . From the above results, it is clear that the SBS reflectivity for a multi-mode pump with a large number of longitudinal modes is significantly affected by self-focusing due to the high-intensity spikes. The reflectivity for the multi-mode pump increases with energy up to 6 mJ and decreases strongly thereafter because the breakdown becomes severe, whereas the single-mode pump provides monotonically increasing reflectivity.

Finally, Fig. 6 shows the measured reflectivity of  $\text{CS}_2$ . Of the four liquids examined,  $\text{CS}_2$  has the lowest SBS threshold energy,  $\sim 0.3$  mJ, and the highest reflectivity,  $\sim 95\%$ , in the single-mode pump case because it has the highest steady-state SBS gain (Table 1). On the contrary, the SBS reflectivity for the multi-mode pump is the lowest and almost zero in the entire region. The critical power,  $\sim 20$  kW, for self-focusing is about half of the SBS threshold,  $\sim 40$  kW. Besides,  $\text{CS}_2$  has the longest acoustic lifetime, 6 ns among the liquids used [15], which is comparable to the pulse-width ( $6 \sim 8$  ns) of the pump beam. As we already mentioned, if the temporal fluctuations of the pump pulse are fast relative to the acoustic phonon lifetime, the acoustic waves do not have time to build up. As a result,  $\text{CS}_2$  has a very low breakdown threshold,  $< 0.3$  mJ, which can account for the almost zero reflectivity observed. We have observed that optical breakdown results in a filament in the case of a very weak pump energy close to the SBS threshold. It is noted that stimulated Raman scattering (SRS) may also be responsible for the low reflectivity because  $\text{CS}_2$  has a high SRS gain [8].

#### IV. CONCLUSION

We have investigated SBS reflectivity for a multi-mode pump with a large number of longitudinal modes, and compared the results with those for a single-mode pump. We used four kinds of liquids with different nonlinear refractive indices and Brillouin line-widths. We found that in all the cases except  $\text{CS}_2$ , the SBS reflectivity for the multi-mode pump increased rapidly and then decreased slowly as the pump energy increased although the SBS gain was as high as that for the single-mode pump. For the case of  $\text{CS}_2$ , the reflectivity was generally very low although the nature of variation was similar to those for the other liquids. On the contrary, the single-mode pump provided a stable and high reflectivity. This is because the multi-mode pulse, consisting of a large number of longitudinal modes, has high-intensity spikes created by

mode beating, which have enough power to induce optical breakdown and nonlinear effects, such as self-focusing and SRS. Especially, we found that the SBS reflectivity using a multi-mode pump was significantly affected by self-focusing which enhanced SBS even at powers lower than the SBS threshold and suppressed SBS by generating strong optical breakdown at high pump energies.

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