

Stimulated Brillouin Scattering in SF₆ with a Free-Running XeCl Laser as Pump

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Abstract. A XeCl laser system without dispersive elements is used to investigate near threshold reflectivity and phaseconjugation (PC) fidelity of stimulated Brillouin scattering (SBS) mirrors with SF₆ as the active medium. Using different focal-length lenses to focus the broadband laser radiation into the Brillouin medium, it was found that at threshold the effective interaction length for SBS is equal to the confocal parameter and that the SBS gain is equal to its steady-state value for monochromatic pumping. High PC-fidelity values ranging from 0.8–1.0 were found under most of the experimental conditions investigated.

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The development of broadband stimulated Brillouin scattering (SBS) reflectors to compensate phase distortions in optically distorted amplifiers without limiting the bandwidth of the oscillating radiation has received great interest over recent years [1–4]. In many laser applications the use of large spectral-width laser radiation helps to suppress deleterious parametric instabilities [5], and not all lasers can easily be made to operate efficiently in a narrow band [6]. Therefore, there exists a need to investigate experimentally phase conjugation by SBS using broadband pumping.

Recently, we have carried out some experimental studies on SBS by broadband pumping [7, 8]. A review of theoretical and experimental works on broadband SBS is also given in these papers and in the references cited therein. The reflectivity and the phase-conjugation fidelity of SBS mirrors versus pump intensity at different pump bandwidths $\Delta \nu_{\rm p}$ were investigated in [7]. Methanol and cyclohexane were used as active media. It was found that by increasing $\Delta \nu_{\rm p}$ from 37 GHz to 75 GHz the threshold intensity for SBS remained unchanged but the fidelity and the reflectivity of the SBS mirrors were quite dependent on pump inten-

sity and spectral width. A peak energy reflectivity of 90% was obtained at the highest value of $I_p = 50 \text{ GW/cm}^2 \text{ em-}$ ployed for the pump intensity by using a pump beam with a bandwidth of $\Delta \nu_{\rm p} = 37 \, {\rm GHz}$ whereas only a peak reflectivity of 60% was obtained at $I_{\rm p} = 50 \, {\rm GW/cm^2}$ with the $\Delta \nu_{\rm p} = 75 \, {\rm GHz}$ pump beam. High fidelity values of (1.0– (0.8) were measured near threshold but a decrease in fidelity with increasing $I_{\rm p}$ was also observed. Moreover, the fidelity decrease with $I_{\rm p}$ was faster for the $\Delta \nu_{\rm p} = 75 \, \text{GHz}$ pump beam. In accordance with such experimental results it was shown in [8] that a liquid SBS reflector is effective in correcting the spatial aberrations of laser radiation originating in an amplifier if the pump intensity in the focused beam is kept below 1 GW/cm². It was shown that long focal-length lenses should be used in the SBS mirror to obtain lower pump intensities and thereby output beams with a higher phase-conjugate fidelity.

In [2] a dual spectral-line pump whose line separation was variable from zero to 4.4 GHz was used to measure the SBS reflectivity. A significant drop in reflectivity was observed when the interaction length l_i for SBS was longer than the pump coherence length $l_{\rm c}$. This drop in reflectivity was attributed to the fact that whenever $l_i < l_c$, the effective interaction length for SBS must be set equal to the pump coherence length. The parameters which govern the effective interaction length at threshold for phase conjugation due to SBS were also investigated experimentally by Munch et al. [4] using a high power, near diffraction-limited ruby laser whose coherence length could be varied from $l_c = 8 \text{ cm}$ to $l_c = 1$ cm. The authors of [4] found the effective interaction length to be the shorter of the following parameters: the cell length, 3 times the coherence length or 5 times the Rayleigh range of the input laser radiation. They also observed that the fidelity of the Brillouin-backscattered radiation depends strongly on the pump coherence length.

In the present experimental work a XeCl laser system without dispersive elements was used to investigate reflectivity and phase-conjugation fidelity of SBS mirrors near threshold. By focusing the laser radiation into the Brillouin medium with different focal-length lenses, the gain and interaction length for SBS by broadband pumping is estimated.

Experimental Apparatus

A free-running oscillator-amplifier XeCl laser system similar to the one described in [8] was used in the present experimental work. In particular, a plane-plane cavity composed of an aluminized mirror and of a quartz flat with two intracavity spatial filters of 1 mm diameter, was applied to the oscillator to obtain a nearly diffraction-limited laser beam. The expanded oscillator beam was then injected into the amplifier which provided a laser pulse of 13 mm diameter, 7 mJ energy, 11 ns duration (full width at half maximum, FWHM), and a divergence angle of $\vartheta_{\rm p}=0.18\,{\rm mrad}$ containing 85% of the total pulse energy. This value is 3 times larger than the diffraction-limited value of $\vartheta_{\rm d} = 2.44\lambda/D = 0.06$ mrad, where λ is the laser wavelength and D the beam diameter. A typical spectrum of the pump radiation monitored by a Jobin-Yvon THR 1500 spectrograph is shown in Fig. 1. The main peaks are due to the two strongest 0-1 and 0-2 electronic transitions [9] at 3079.8 Å and at 3081.9 Å. Besides these, quite weak 0-0 and 0-3 emissions at 3077.2 Å and at 3084.5 Å were also present, which are not shown in Fig. 1. The pump radiation spectrum extends then over a width of $\varDelta\lambda_{\rm p}\simeq 7.8~{\rm \AA}.$

Thus, a non-diffraction-limited laser beam of quite a large bandwidth was used in our measurements.

Due to the high-gain [10, 11] and the good transparency throughout the ultraviolet region of the spectrum, SF_6 was used as the Brillouin medium whereby a pressure of 1.6 MPa was chosen. Gas of 99.9% purity was provided by Messer-Griesheim (Hoechst, Italia). The stainless stell SF_6 cell sealed with tilted (5°) quartz windows was 50 cm long.



Fig. 1. Spectrum of the XeCl-laser radiation pumping the SBS mirror



Fig. 2. Schematic of the experimental setup

For simultaneously measuring SBS reflectivity and farfield fidelity versus pump energy, the experimental setup schematically shown in Fig. 2 was used. The neutral density (ND) filters allow to vary the pump beam energy. The lens $L_{\rm B}$ with a focal length $f_{\rm L}$ ranging from 15 cm to 50 cm focus the pump beam into the 50 cm long SBS cell containing SF_6 . Care was taken to direct the pump beam at an angle into the cell to avoid the collection of the light rectro-reflected by the SF_6 cell window and by the lens surfaces. Moreover, the pump beam was focused into the cell at about 10 cm from the cell entrance window. For the reflectivity measurements the Gen-Tec pyroelectric detectors D1 and D2 sample the pump and the Brillouin backscattered energy taken through the beam splitter B, respectively. The phase-conjugation (PC) fidelity of the backscattered radiation was determined by using the energy-in-the-bucket technique as reported in [12]. Here, the energy fraction ε of the Stokes radiation contained within the same divergence angle as that of the pump beam $\vartheta_{\rm p} = 180\,\mu {\rm rad}$ (85% of the total energy is within $\vartheta_{\rm p}$ as mentioned earlier) is measured firstly. The PC fidelity is then defined by the ratio $\varepsilon/0.85$. The analysis of the Stokes beam was carried out by focusing it with a 5 m focal-length lens and by measuring the energy in the focal plane of the lens one time without an aperture the other time with an aperture having a diameter of $\vartheta_{\rm p} f = 0.18 \times 10^{-3} \cdot 5 \,{\rm m} = 0.9 \,{\rm mm}.$

Experimental Results and Discussion

Figures 3 and 4 show the SBS mirror reflectivity versus the pump beam energy E_p and the focal beam intensity I_p for the lenses with different focal lengths. I_p has been calculated using the following expression

$$I_{\rm p} = \frac{E_{\rm p}}{\Delta t_{\rm p} \pi (f_{\rm L} \vartheta_{\rm p}/2)^2} , \qquad (1)$$

where $\Delta t_{\rm p} \simeq 11$ ns is the pulse length of the pump beam measured by taking radiation through the beam splitter B. The reflectivity fluctuations are indicative of the random character of the noise which seeds the SBS process and are not due to measurement noise.

It can be seen from Figs. 3 and 4 that the threshold pump energy E_p^{th} is independent on the focal length f_{L} of the SBS mirror lens. The growth of the SBS mirror reflectivity starts at about 3.3–3.5 mJ for all values of f_{L} ranging from 15 cm up to 50 cm. In fact, if l_i is the interaction lenght, the SBS signal is expected to depend on the product $I_p l_i \propto I_p f_{\text{L}}^2$. Because of (1), this product is independent on f_{L} in the absence of competitive nonlinear processes. In accordance to that, a strong dependence of the threshold pump intensity I_p^{th} on f_{L} can be clearly seen from the Figs. 3 and 4. In particular, Fig. 5 shows that I_p^{th} increases linearly with $1/f_{\text{L}}^2$. Then, with regard to Fig. 5 one can say that $l_i = \text{const.} f_{\text{L}}^2$. We believe that the following arguments allow to estimate the value of the constant in this equation.

SBS starting from noise under transient conditions is considered to occur when the exponential gain, approximately given [2] by

$$G \approx 2(g_{\rm B} l_{\rm i} I_{\rm p}^{\rm th} \Delta t_{\rm p} / \tau_{\rm B})^{1/2} , \qquad (2)$$



Fig. 4. SBS energy reflectivity vs pump intensity in the focal plane for $f_{\rm L} = 30$ cm, 37.5 cm, and 50 cm



Fig. 5. Threshold pump intensity as a function of the focal length of the SBS-mirror lens

reaches values ranging from 25 to 30, where $g_{\rm B}$ is the steady state SBS gain coefficient which depends only on the kind of Brillouin medium used and $\tau_{\rm B}$ is its pertinent acoustic decay time. Converting the conditions investigated in [9] to our pressure of 1.6 MPa and our wavelength of 308 nm one finds $\tau_{\rm B} = 2.2$ ns for SF₆, which is of the order of our pumppulse length of 11 ns. This supports the use of the threshold condition (2) to our situation.

By assuming the interaction length l_i equal to the confocal parameter z_0 given by $(2\pi/\lambda)(f_L\vartheta_p/2)^2$ [13] and by fitting the experimental results of Fig. 5 with (2), taking G = 25, one finds $g_B \cong 1.5$ cm/GW which is in good agreement with the calculated value reported in [14] for monochromatic pumping. Our experimental results seem thus to indicate that at threshold the assumption $l_i = z_0$ leads to the conclusion that there is no difference between broadband and monochromatic pumping, as far as the gain coefficient is concerned. This is in agreement with theoretical considerations as outlined in [15].

It is worth mentioning that by changing $f_{\rm L}$ from 15 cm to 50 cm the value of the confocal parameter and thus also $l_{\rm i}$, was varied from 3.7 mm to 41 mm, respectively. Even by considering $l_{\rm c} \simeq 2.4$ mm corresponding to $\Delta \lambda \simeq 40$ pm as "effective pump coherence length" one finds that the interaction length was then changed from about 1.5 $l_{\rm c}$ to about 16 $l_{\rm c}$ under our experimental conditions. Then, despite of the conclusions of [4], we have observed that at threshold the effective interaction length is not limited by the pump coherence length to be approximately equal to 3 $l_{\rm c}$.

Furthermore, Fig. 3 clearly points out the strong dependence of the energy reflectivity R on the pump intensity I_p when short focal-length lenses are used to focus the laser radiation into the Brillouin medium. After a strong increase the reflectivity reaches a maximum and then decreases slowly. The two cases shown differ from each other in that the maxima are not equal and occur at different values of $I_p: R = 12\%$ for $I_p = 60 \text{ GW/cm}^2$ and $f_L = 15 \text{ cm}$ versus R = 17% for $I_p = 30 \text{ GW/cm}^2$ and $f_L = 20 \text{ cm}$. However, when the pump radiation is focused with a 10 cm focal-length lens, reflectivities not larger than 3% could be measured.

These last findings are consistent with those reported in [16], where a 2 cm focal-length lens was used to focus broadband XeCl-laser pulses of 40 mJ energy and 15 ns duration into SF₆. No coherent backscattering was observed, even by varying the SF₆ pressure from 0.1 MPa up to 2.0 MPa.

The onset of nonlinear processes such as stimulated Raman scattering, self-focusing, optical heating which compete with SBS in the focal region depends on the pump beam intensity [10, 12]. In particular, it is believed that the use of pump radiation with a large bandwidth may facilitate the onset of these competitive processes [3]. It could thus be that the quite low reflectivities measured when using short focal-length lenses and the reflectivity decrease at high pump intensities observed with $f_{\rm L} = 15$ cm and $f_{\rm L} = 20$ cm are associated with the competition between SBS and these non-linear processes.

The laser radiation should thus be focused into the Brillouin medium with a long focal-length lens to have low pump intensities and to reduce the threshold for the onset of competitive nonlinear processes. The experimental results of Fig. 4 suggest that, under our experimental conditions, higher reflectivities appear to be attainable by the SBS mirror at $f_L \ge 30$ cm by increasing the pump energy. Unfortunately, our laser was not strong enough to demonstrate this tendency for $f_L = 37.5$ cm and 50 cm, respectively.

Similar experimental results have been obtained by using liquids as active media [7, 8]. However, it has been observed in [8] that the focal length of the SBS-mirror lens should not be much longer than 20 cm to obtain relatively high mirror reflectivities. It is worth mentioning that with methanol as a Brillouin medium it has been observed that the SBS-mirror reflectivity starts at a threshold pump energy of 0.2–0.3 mJ both with the $\Delta \nu_{\rm p} = 37$ GHz [7] and with the free-running pump beam [8]. Such values of $E_{\rm p}^{\rm th}$ are about 10 times



Fig. 6. Phase-conjugation fidelity vs the pump intensity for $f_{\rm L} = 50$ cm (\Box); $f_{\rm L} = 37.5$ cm (\blacktriangle); $f_{\rm L} = 30$ cm (\circ); $f_{\rm L} = 20$ cm (\triangle); $f_{\rm L} = 15$ cm (\bullet)



Fig. 7a, b. Time evolution a of the pump pulse and b of the Brillouinbackscattered pulse

smaller than those measured for SF₆ but, the SBS gain for methanol, $g_{\rm B} = 13$ cm/GW, is about 10 times larger than that for SF₆.

Recently, the SBS reflectivity of 26 organic liquids using an unmodified broadband XeCl laser was investigated in [17]. Divergence and focusing conditions were demonstrated as being important to achieve sufficient coherence in the scattering area.

Figure 6 shows the phase-conjugation fidelity measurements versus I_p for all the different values of f_L . The PC-fidelity values greater than one are only due to an artifact of the energy-in-the-bucket technique, as it was already stated in [12]. High-fidelity values ranging from 0.8–1 were found under most of the experimental conditions investigated. Only at the highest pump intensities a decrease of the average PC-fidelity value occurred as one should expect due to the onset of competitive nonlinear processes [10]. A similar variation of the fidelity versus I_p was also observed when using methanol as the Brillouin medium [7, 8]. In particular, high-fidelity values were only obtained at pump intensities close to threshold.

To within an accuracy of $\sim 0.5 \text{ cm}^{-1}$, which is due to the spectrograph used, the spectrum of the Brillouinbackscattered radiation turned out to be equal to that of the pump radiation. The Brillouin-frequency shift which is $\simeq 0.03 \text{ cm}^{-1}$ [8] in our case is lower than the resolution limit of our measurement system and could thus not be detected.

We have also measured the time evolution of the pump and the backscattered pulses by recording the radiation through the beam splitter B. Figure 7 shows a typical example.

Under all experimental conditions chosen, the backscattered pulses had a duration of \sim 7.5 ns. Then, since the pump pulse was 11 ns long, a Stokes pulse shortening of about 30% was obtained (see Fig. 7). In particular, with respect to the focus in the SBS cell, the backscattered pulse appeared about 1–2 ns after the pump pulse. Then, with respect to the pump pulse both the leading and the falling edge of the Stokes pulse appear to be somewhat shortened.

Conclusions

A broadband XeCl laser was used to investigate nearthreshold SBS in SF₆ versus pump intensity. A fast decrease of reflectivity at high pump intensities has been observed by focusing the laser radiation with short focal-length lenses $(f_{\rm L} \leq 20 \text{ cm})$. It is believed that under our experimental conditions the onset of nonlinear processes which compete with SBS has prevented the occurrence of high reflectivities at high pump intensities. Nevertheless, we have demonstrated that high-quality phase-conjugate reflections can be obtained by SBS in SF₆ even with broadband pumping.

Thus, broadband SBS mirrors using SF_6 as active medium can be used for building phase-conjugating mirrors which, as it is well known, are applied for compensating phase distortions in high-power laser systems [15].

It has also been shown that near threshold the SBS gain is close to its steady-state value for monochromatic pumping and that the effective interaction length is of the order of the focused beam confocal parameter.

We believe that our experimental results in combination with those obtained for liquid SBS media will help to understand SBS better when using a broadband pump.

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