Pulse compression to one-tenth of phonon lifetime using quasi-steady-state stimulated Brillouin scattering

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Abstract: A new stimulated Brillouin scattering (SBS) compression mechanism, quasi-steady-state SBS compression, in which the compression limit is one-tenth of the phonon lifetime with a high energy efficiency, is proposed and practically realized in this study. The feasibility of this approach is demonstrated experimentally, in which a compression of \(0.36\tau_B\) with an energy efficiency of 65% is achieved in a 3M Fluorinert Electronic Liquid FC-3283 and a compression output of \(0.12\tau_B\) (near-compression-limited) with an energy efficiency above 40% is obtained in acetone when the phonon lifetime to leading-edge to ratio is greater than 10. This ratio is identified experimentally as the key parameter in quasi-steady-state SBS compression. This work provides a practical approach to reliably generating one-tenth-phonon-lifetime pulses by quasi-steady-state SBS compression.

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OCIS codes: (190.0190) Nonlinear optics; (290.5900) Scattering, stimulated Brillouin; (320.5520) Pulse compression.

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1. Introduction

In recent years, there has been considerable interest in the development of ultrashort-pulse and high-power lasers for application to high harmonic generation with high spectral purity for atomic and molecular spectroscopy [1, 2], efficient pump sources for optical parametric amplifiers [3, 4], and high spatial resolutions in LIDAR Thomson scattering diagnostics [5]. Widely applied in the field of high-energy sub-nanosecond generation [6], pulse compression by stimulated Brillouin scattering (SBS) is a promising way to generate ultrashort and high-intensity pulses with good spatial quality [7]. Theoretically, the limit of steady-state SBS pulse compression is set by the phonon lifetime $\tau_B$ on the timescale of hundreds of picoseconds, whereas the limit of transient SBS is the half-cycle gain time $\tau_a$ of the order of picoseconds [8]. Benefiting from the substantial progress in both the configuration [9–12] and SBS active medium [13–17], several studies have achieved near-compression-limited output with a high energy and efficiency in the steady-state regime [18, 19]. It has widely been used in commercial laser and large laser systems to obtain pulses with widths of hundreds of picoseconds [20, 21]. In contrast, output pulse durations much shorter than the phonon lifetime duration can be expected when the pump pulses are shorter than the phonon lifetime of the Brillouin medium $\tau_B$, namely, in the transient regime. A detailed discussion of this technique was reported by Velchev et al., and $0.54\tau_B$ was achieved in their experiment [8]. Although the physical limit of the transient SBS pulse compressor is the acoustical half-cycle duration, it is not clear whether this can occur in real systems. Low output energy and energy conversion efficiencies, caused by the strong reduction of gain in the transient region, also hinder its practical application.

In the steady-state regime, although further compression can be realized by a medium with a short phonon lifetime, femtosecond or picosecond lasers require unrealistically high densities of the SBS medium [7]. The generation of sub-phonon-lifetime pulse compression with high energy efficiencies has become an important research topic. Experimentally, to push the SBS technique to realize further compression, many studies have identified and optimized critical parameters. Xu et al. suggested that the effective interaction length is one of the key factors for achieving sub-phonon-lifetime pulse compression and demonstrated an output duration of $0.5\tau_B$ using approximately 100 mJ and the full-length at half-maximum (FWHM) of the input Gaussian pulses [22]. Neshev et al. found that the spatial beam profiles of the incident pump beam had an influence on the minimum output pulse duration [23]. Yoshida demonstrated a compression result of $0.8\tau_B$ with an 80% energy efficiency by eliminating the negative effect of the transverse beam profile [6]. In the physical mechanism, Yuan et al. developed a theoretical model to demonstrate that fluctuations in the Stokes signal and occurrence position are key factors for obtaining the narrowest Stokes pulses. They achieved an output duration of $0.7\tau_B$ in the optimum Stokes occurrence position [24]. Unless injection-seeded, the backward Stokes pulse is naturally random because SBS is generated from thermally excited density fluctuations, which leads to a spread in the pulse duration and peak intensity [25]. In practice, the SBS generator is an essential part of current SBS devices [9–12], including a real seed injection setup [19]. Several proposals have been made to suppress and fix the fluctuations, such as a non-focusing SBS scheme [26] and interferometric SBS scheme [27–30]. Because of the complex design and stringent requirements of the pump shape, the non-focusing scheme is better suited for large laser facilities. As the complexity and losses of optical surfaces are greatly reduced, the interferometric SBS scheme has more advantages in small device and commercial laser applications. Despite certain breakthroughs in both theory and practice, the optimization of steady-state SBS compression can only provide a small improvement.
Both steady-state and transient SBS pulse compression suffer from great limitations in the minimum Stokes duration or energy conversion efficiency. Quasi-steady-state SBS is a special case that it belongs to steady state with initial duration longer than phonon lifetime, by definition, but it includes transient process in the generation or amplification (normally in the process of Stokes generation). We use the term “quasi-steady state” to indicate the special steady-state SBS compression including transient effects in this study. Integrating the advantages of both the steady-state and transient regime, quasi-steady-state SBS pulse compression can alleviate such constraints. It has been proved that the gain of SBS is reduced only slightly in the quasi-steady-state region [31]. Theoretical simulations predict that the formation of pulses with a duration of $0.1\tau_B$, characterized by high conversion efficiencies (~50-70%), is possible if the leading edges of the initiating pulses are sufficiently steep compared to the phonon lifetime [32]. However, the analysis of approximate solutions allows us to only come to certain qualitative conclusions, and it is still a challenge to determine how to practically implement these solutions.

In this paper, we present an implementation scheme for the realization of quasi-steady-state SBS compression by choosing pump pulses with a leading-edge shorter and duration longer than the phonon lifetime. The phonon lifetime to leading-edge ratio is identified experimentally as the key parameter in quasi-steady-state SBS compression. Using the combination of an interferometric scheme and step pump pulse, we demonstrated a compression of $0.36\tau_B$ with an energy efficiency of 65% in a 3M Fluorinert Electronic Liquid FC-3283. By increasing the phonon lifetime to leading-edge ratio, the compression is enhanced gradually and a (near-compression-limited) compression output of $0.12\tau_B$ with an energy efficiency above 40% in acetone eventually. To the best of our knowledge, this is the first time a pulse compression to one-tenth of the phonon lifetime has been experimentally achieved with a high energy conversion efficiency.

2. Experimental setup

Quasi-steady-state SBS is a transition state from a steady-state to transient regime [31]. In practice, it can be implemented with steep-leading and long pump pulses to establish an acoustic field in the transient regime and amplify Stokes radiation under a steady state. The leading edge of the pump pulse is crucial in achieving a minimum output duration, and the tailing edge largely determines the energy efficiency. Therefore, the step pulse, which can effectively be achieved using an SBS waveform generator [33], is considered an optimum pump waveform. The sharpness of the leading edge is relative to the phonon lifetime of the medium. It is difficult to quantitatively estimate the sharpness of the pump leading-edge in the theoretical model. In contrast, to avoid the impact of pump power variation, we can perform SBS pulse compression in an experiment using the same pump pulses in a different SBS medium (different phonon lifetimes) to confirm the optimum ratio of the phonon lifetime to leading-edge. Here, acetone and 3M Fluorinert Electronic Liquids FC-3283, FC-75, FC-72 whose SBS parameters are listed in Table 1, are selected as the SBS medium.

| Table 1. Parameters of SBS Medium used in Experiments |
|-----------------|----------------|----------------|----------------|----------------|----------------|
| SBS medium      | HT-230 [34]   | FC-3283        | FC-75 [34]     | FC-72 [34]     | Acetone [35]   |
| Wavelength $\lambda$ (nm) | 1064          | 1064           | 1064           | 1064           | 1064           |
| Refractive index $n$      | 1.283         | 1.266          | 1.276          | 1.251          | 0.792          |
| Phonon lifetime $\tau_B$ (ns) | 0.1           | 0.6            | 0.9            | 1.2            | 2.67           |
| $\tau_p = 1 / \Gamma_p$ (ns) |               |                |                |                |                |
| Brillouin gain factor $g_{\text{th}}$ (cm/GW) | 3.8           | 4.5            | 5              | 6              | 15.8           |
A schematic of the experimental setup is illustrated in Fig. 1. The primary pump source is a custom-built Q-switched Nd:YAG laser delivering single-longitudinal-mode laser pulses with a pulse duration of 8 ns and wavelength of 1064 nm. The laser resonator is capable of supplying an output energy of approximately 7 mJ at a repetition rate of 1 Hz. An isolator, comprised of a Faraday rotator (FR), polarizer P1, and a half-wave plate ½, avoids the backscattering light to the laser resonator. The light beam is subsequently expanded to a diameter of 5 mm through a beam expander using a system of lenses L1 and L2. The linearly polarized beam passes through a polarization beam-splitter P2 and is amplified by a flashlamp-pumped Nd:YAG amplifier to an energy greater than 30 mJ. The amplified beam is focused by a plano-convex lens L3 (f = 300 mm) into the SBS waveform generator, filled with HT-230, to create step pulses propagating in the backward direction. After passing through a quarter-wave plate ¼ and amplifier twice, the step pulses are reflected by polarizer P2. The output step shape pulse with a 253 ± 22-ps leading-edge is depicted in Fig. 2.
An interferometric SBS scheme is utilized for quasi-steady-state SBS compression with a step pulse pump. In contrast to the Gaussian pump via a single-cell scheme, this combination does not suffer from the impact of varying effective interaction length. Therefore, the optimum spatial length of the pulse is \((c/2n)\tau_p\), where \(c\) is the speed of light, \(\tau_p\) is the pulse duration (FWHM), and \(n\) is the refractive index of the SBS medium. Transmitted through the 40-cm SBS cell, the circularly polarized laser beam is backward focused by a concave mirror with a focal length of 30 cm. At the focal region, electromagnetic standing wave, which arises from the interferences between the main beam and counter-propagating beam, can induce self-generated periodic density modulation which ignites the Brillouin grating. The temporal SBS compressor provide stable output, and the pulse compression and phase matching mechanism have been described in detail in the literature [27–30]. After the interaction, the polarization of the Stokes pulse is changed by quarter-wave plate \(\lambda/4\), which allows them to be reflected by \(P_3\).

The optical pulse duration characteristics are measured by a fast phototube (UPD-40-UVIR-D, Alphalas GmbH, Germany; rise time < 40 ps) combined with a digital oscilloscope (DPO71254C, Tektronix, USA; bandwidth: 12.5 GHz; sampling rate: 100 Gsamples/s). The laser energy is recorded by a laser energy meter (PE50DIF-ER, Ophir Optronics, Israel).
3. Experimental results

![Graph showing pulse duration evolution of compressed pulses across the entire beam with respect to input energy in FC-3283.](image)

The pulse duration evolution with respect to input energy in FC-3283 is presented in Fig. 3. The phonon lifetime is over twice \(2.37 \times \tau\) the rising time of the pump pulse. The value of each point is the average of 50 pulses. Before the SBS process reaches the gain saturation region, the output Stokes pulse duration narrows rapidly as the pump energy increases. When the SBS process is deep within the gain saturation region, the output pulse duration decreases slowly and tends to a particular value. It should be emphasized that the Stokes pulses obtained here are free of tailing modulation and do not broaden at high pump intensities, as observed in Fig. 3. The SBS in the interferometric scheme has an exceptional mechanism, in which the phase and generation position of the SBS wave are fixed via a weak periodic density modulation in the focal region inside the SBS cell by means of a standing wave. The fixed generation position of the Stokes pulses can effectively control the phenomenon of tailing modulation and Stokes duration broadening in the SBS compression process [27–30]. In contrast, the asymmetry of amplification at the front and tail parts of the Stokes pulse is the essence of SBS pulse compression. The step pump shape can maximize the asymmetry of amplification and enhance the coupling effect of the optical fields (laser and Stokes) and acoustic field, leading to further compression. The corresponding average value of the shortest Stokes pulse is 218 ps \(0.36\tau_B\). Although it is a considerable improvement over previous research on sub-phonon lifetime compression, there are still certain disparities upon comparison with the theoretical results.
In quasi-steady-state SBS compression, the minimum output duration depends on the transient effects in the process of Stokes pulse generation. Larger $\tau_B/\tau_{\text{rising}}$ means stronger transient effects. To further reveal the effect of $\tau_B/\tau_{\text{rising}}$ on SBS compression characteristics, FC-75 and FC-72 with phonon lifetime of 0.9 ns ($\tau_B/\tau_{\text{rising}} = 3.6$) and 1.2 ns ($\tau_B/\tau_{\text{rising}} = 4.7$) are used to perform the contrast experiments. The experimentally measured dependence of compressed pulse duration on input energy with two different SBS medium is presented in Fig. 4. It can be seen that sub-phonon lifetime pulse compression are achieved with both in FC-75 and FC-72. The minimum output duration in FC-75 is $0.34\tau_B$ and that is $0.284\tau_B$ in FC-72. Combining with the output duration $0.36\tau_B$ obtained in FC-3282, it is clear that further compression can be obtain in sub-phonon lifetime compression as the ratio $\tau_B/\tau_{\text{rising}}$ increases. The compression can be further improved by increasing $\tau_B/\tau_{\text{rising}}$.

![Figure 4](image1.png)

**Fig. 4.** Pulse duration evolution of Stokes with respect to input energy. (a) The measurements of Stokes duration in FC-75 with phonon lifetime of 0.9 ns and the corresponding $\tau_B/\tau_{\text{rising}}$ is 3.6. (b) The measurements of Stokes duration in FC-72 with phonon lifetime of 1.2 ns and the corresponding $\tau_B/\tau_{\text{rising}}$ is 4.7.

Acetone with a longer phonon lifetime (2.67 ns) is chosen to further confirm the inference. The measured pulse duration of acetone is plotted as a function of pump pulse energy in Fig. 5. The results are similar to those obtained with FC-3283, FC-75 and FC-72. It can be seen that the duration of the Stokes pulse decreases more rapidly and the saturation is about 325 ps ($\sim 0.12\tau_B$). As expected, for the same input pump energy, a higher phonon lifetime to leading-edge ratio ($\sim 10.55$) can realize further compression. It holds that the further increase in the ratio (above 10) may lead to the transfer of energy back from the Stokes to the pump pulse [36]. Compared with steady-state SBS, the acoustic wave field
establishment of quasi-steady-state SBS is in the region of transient with shorter time, which is favorable for improving SBS compression [33]. It should be noted that the quasi-steady-state SBS is not contrary to the well-known fact that, under the same conditions, shorter compressed pulses can be obtained if a SBS medium with shorter phonon lifetime was used. For a fixed value $\tau_B/\text{rising time}$, shorter compressed pulses can be obtained using SBS medium with shorter phonon lifetime. The SBS medium we selected have appropriate representativeness for commonly used SBS medium. This method offers new possibilities to obtain ultra-short pulses using suitable ratio $(\tau_B/\text{rising time})$ and SBS medium with short phonon lifetime, particularly the other heavy fluorocarbons $\text{C}_N\text{F}_{2N+2}$.

![Fig. 6. Output energy (black points) and energy conversion efficiency (blue points) of Stokes pulses versus input pump energy correspond to (a) FC-3283, (b) FC-75, (c) FC-72 and (d) acetone, respectively.](image)

The output energy and corresponding energy conversion efficiency versus pump energy are presented in Fig. 6. The output energy of all media increase monotonically and the energy conversion efficiency of the SBS tends to a saturation condition at high energy after a rapid increase at low energy. Compared with transient SBS, the Stokes pulses are amplified in steady-state, which is beneficial to obtain high energy conversion efficiency. With the comparison of the curves of the four media, the energy characteristics of the quasi-steady-state SBS is significantly different from the steady-state SBS. The SBS gain coefficient of FC-3283, FC-75, FC-72 and acetone are 4.5 5, 6 and 15.8 cm/GW, respectively. Lower efficiencies are achieved with higher gain coefficients. The maximum SBS efficiency decreases with the increasing of $\tau_B/\text{rising time}$. The increase in $\tau_B/\text{rising time}$ implies a further transition toward transient SBS, and the SBS energy conversion efficiency, influenced by transient effects, decreases consistently. A greater value of $\tau_B/\text{rising time}$ to a certain extent, may imply a lower energy conversion efficiency. In contrast, a considerable amount of previous research indicates that although the SBS pulse compression is limited by the phonon lifetime when the pump pulse duration is on the level of nanoseconds, the realization of near-compression-limited output is restricted by the gain coefficient of the SBS medium [6, 35], whether in the steady state or quasi-steady state. Here, the significant gain decrease implies
that the compression performance improvement of acetone contributes to the increase in $\tau_B/\text{rising time}$ instead of a higher gain coefficient.

4. Conclusions

In summary, we have demonstrated an approach to practically realize quasi-steady-state SBS compression, and we have achieved a (near-compression-limited) compression output of $0.12\tau_B$ with an energy efficiency above 40% when the phonon lifetime to leading-edge ratio is greater than 10. These results suggest that quasi-steady-state SBS compression is an extremely effective way to generate high-energy ultrashort pulses from nanosecond pulses. In addition, this mechanism can work in all SBS media and SBS compressor schemes. This mechanism can further push the compression performance of SBS pulse compression, which is a mature and effective technique for generating high-energy sub-nanosecond pulses with good beam quality. It should be noted that some optical components we used were uncoated, which makes the SBS far from the theoretical conversion efficiency. Ultrashort pulses can be obtained if an SBS medium with a shorter phonon lifetime is used or the input pulses are compressed at shorter wavelengths as $\tau_B$ is proportional to $\lambda^2$ [38]. Meanwhile, the waveform generator can be further optimized to an expanded application range. A combination of fiber lasers and fiber intensity modulators driven by an arbitrary waveform generator can generate step pulses with different steepness values to match different SBS media and pump wavelengths. The quasi-steady-state SBS compression can be a feasible technique for generating single-frequency, picosecond pulses with high efficiencies by the further optimization and improvement of experimental conditions.

Funding

National Natural Science Foundation of China for Excellent Young Scholars (61622501)

Acknowledgments

The authors would like to acknowledge technical support of Hengkang Zhang from the State Key Laboratory of Precision Measurement Technology and Instruments, Department of Precision Instrument, Tsinghua University.