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SRS converter–compressor of femtosecond ytterbium laser pulses

A.V. Konyashchenko, L.L. Losev, V.S. Pazyuk

Abstract. **Stimulated Raman scattering (SRS) conversion of femtosecond pulses of an ytterbium laser at a wavelength of 1.03 μm is performed in compressed hydrogen under conditions of nonlinear phase modulation. Temporal compression of frequency-chirped Stokes pulses is carried out, as a result of which 400-fs laser pulses are converted into 27-fs Stokes pulses with a wavelength of 1.8 μm. An average Stokes power of 0.6 W is implemented at an average laser power of 6.3 W and a pulse repetition rate of 20 kHz.**

Keywords: *stimulated Raman scattering, femtosecond laser, nonlinear phase modulation, pulse compression.*

1. Introduction

It was shown in [1] that frequency conversion of a femtosecond laser pulse by stimulated Raman scattering (SRS) can be performed simultaneously with nonlinear phase modulation of the laser and Stokes waves. These processes give rise to a broadband frequency-modulated (chirped) Stokes pulse at the output of a Raman-active medium. The compensation for the frequency chirp by dispersion optical elements placed after the active medium makes it possible to temporarily compress the Stokes pulse. An SRS conversion of ytterbium laser radiation with a pulse duration of 270 fs into Stokes radiation with a centre wavelength of 1.8 μm was performed in compressed hydrogen. A positively chirped Stokes pulse was compressed to a duration of 35 fs in optical elements made of fused silica, which has a negative second-order dispersion in the Stokes wavelength range. The energy conversion efficiency reached 6% [1]. The application of this fairly simple scheme of a frequency converter–compressor in combination with a femtosecond ytterbium laser (one of the most developed and widely used modern lasers) is a promising approach for designing a source of femtosecond (with a duration of several light-field cycles) mid-IR pulses with high average and peak powers. In particular, these sources with a gigawatt peak power can be used in experiments on generation of higher harmonics and attosecond pulses in gas media [2].

In this paper, we report the results of a detailed study of the SRS processes and nonlinear phase modulation of femtosecond ytterbium laser radiation in compressed hydrogen, which was aimed at generating maximally short pulses with a maximally high power at a centre wavelength of 1.8 μm.

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2. Experimental setup

An optical scheme of the experimental setup is presented in Fig. 1. A femtosecond ytterbium laser system TETA-10 (Avesta) was used to pump the SRS converter. The laser system contains a master fiber generator of femtosecond pulses and a regenerative amplifier for chirped-pulse amplification. The pulse duration was reduced to 225 fs after the grating compressor placed at the regenerative amplifier output. The maximum values of the single pulse energy, average power, and pulse repetition rate (PRR) were, respectively, 400 μJ, 10 W, and 200 kHz. The radiation spectral width was 7 nm, and the centre wavelength was 1030 nm. This laser system made it possible to vary the output pulse duration from 225 fs to 10 ps by means of a programmable change in the distance between the temporal compressor gratings at the regenerative amplifier output. The diameter of the output laser beam with a Gaussian intensity distribution was 3 mm at a level of e^{-2}.

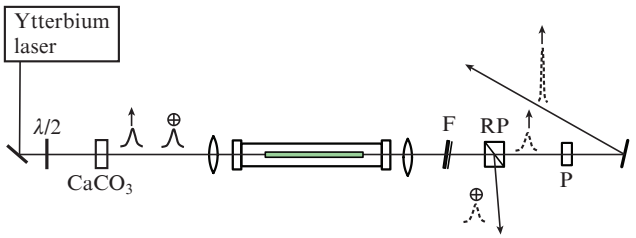

Figure 1. Optical scheme of the experimental setup: ($\lambda/2$) half-wave phase plate; ($CaCO_3$) calcite crystal; (F) broadband filter; (RP) Rochon prism; (P) fused silica plate.

The choice of the optical scheme of the SRS converter was based on the condition of maximally possible spectral width of the laser pulse formed as a result of nonlinear self-phase modulation in a Raman-active medium. In this case, one would also expect a maximum spectral width of the Stokes pulse during SRS conversion and nonlinear cross-phase modulation and, correspondingly, the minimal Stokes pulse duration after the temporal compression. At fixed values of the laser pulse energy and power (the latter should not exceed the critical self-focusing power in compressed hydrogen), this can

be implemented by varying the pulse duration and shape, for example, by dividing the laser pulse into two pulses having approximately equal energies and twice as short as the initial pulse. The pulse peak power is retained in both cases: one pulse with an energy E and a duration 2τ and two pulses with energies $E/2$ and durations τ. The Raman gain increment does not change, because it is determined by only the pulse energy under time-dependent SRS conditions [3]. Since the spectral width $\Delta\nu$ of a pulse with a duration τ subjected to nonlinear self-phase modulation is given by the expression $\Delta\nu \backsim P/\tau$ [4], where P is the pulse power, it will be two times smaller in the first case (one pulse of duration 2τ) than in the case of two pulses. Moreover, the frequency chirp (equal to the ratio of the spectral width of a pulse to its duration) for the double pulse will be larger by a factor of 4 than for the single pulse. Due to this circumstance, one can use optical elements with a smaller negative dispersion for temporal compression and thus simplify the optical scheme and improve the compressed pulse quality. Based on these considerations, we chose an optical scheme with formation of a double pump pulse for the Raman converter.

The double pump pulse for the SRS converter was obtained by transmitting an ytterbium laser pulse through a 1-cm-thick birefringent calcite ($CaCO_3$) crystal. As a result, there arose two orthogonally polarised pulses separated by 5 ps. The energy ratio for these pulses was varied by rotating a phase half-wave plate $\lambda/2$. The delayed-pulse conversion efficiency was the same as in the case of two pulses with identical polarisations, because under conditions of SRS from vibrational hydrogen levels (which is of scalar type) a delayed pulse is scattered from the polarisability grating formed by the first pulse [5].

The laser beam was focused by a lens with a focal length of 30 cm on the input end face of a 80-cm-long quartz capillary with an inner diameter of 250 μm, placed in a 1-m-long stainless steel chamber. The thickness of fused silica windows was 5 mm. The distance from the capillary input to the chamber input window was 12 cm. The chamber was filled with compressed hydrogen. The optical elements at the converter input (half-wave phase plate, calcite crystal, focusing lens, and chamber input window) had an antireflection coating for the ytterbium laser wavelength.

The Stokes radiation at the chamber output was separated from the pump radiation by a broadband filter F (see Fig. 1). A Rochon prism was used to separate Stokes pulses with orthogonal polarisations. A fused silica plate P was placed after the Rochon prism to perform a temporal compression of chirped Stokes pulse.

The duration of compressed Stokes pulse was measured by an ASF-15 autocorrelator (Avesta). The pulse spectra were recorded using a scanning mid-IR spectrometer ASR-IR-2.6 (Avesta).

3. Energy characteristics of the SRS converter

It was shown in [6] that, beginning with a certain femtosecond pulse duration, the SRS conversion efficiency begins to decrease sharply with pump pulse shortening; this drop is due to the processes of nonlinear phase modulation of laser and Stokes waves. In the case of Raman-active gaseous media, the femtosecond pulse duration corresponding to the onset of conversion efficiency drop is independent of the pressure in the medium. To determine the ytterbium laser pulse duration under conditions of SRS in hydrogen, we measured the conversion efficiency as a function of pulse duration. The measurement results are presented in Fig. 2. A scheme of pumping by a doublet composed of orthogonally polarised pulses with equal energies was applied. The experiment was performed at a hydrogen pressure of 27 atm, a PRR of 10 kHz, and a double pulse energy of 250 μJ. The minimum pulse duration in the double pump pulse was 225 fs. In this case the maximum peak power under the experimental conditions reached 0.56 GW, while the critical self-focusing power for ytterbium laser beam in hydrogen (P_{cr}[GW] = 20/p_H[atm] [7], where p_H is the hydrogen pressure) was higher: 0.74 GW.

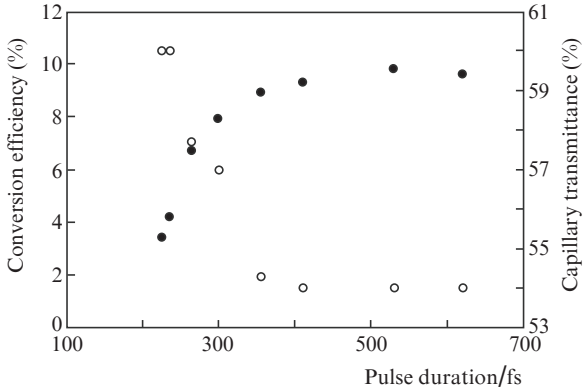

Figure 2. Dependences of the (•) conversion efficiency and (○) capillary transmittance on the single pulse duration in the scheme with double-pulse pumping.

The durations of single pulses in the doublet were changed synchronously by changing the distance between the compressor gratings at the output of the ytterbium laser regenerative amplifier. The pulse duration was increased by means of positive frequency chirping (the instantaneous frequency increases from the pulse leading edge). As can be seen in Fig. 2, the conversion efficiency is saturated at a level of ~9% for pulses longer than 400 fs. At the same time, the delayed Stokes pulse contains more than 90% of energy for the entire range of pump pulse widths under study. This result is in agreement with the estimates reported in [1]. The results of measuring the capillary transmittance also confirmed the influence of phase modulation (rather than self-focusing) on the conversion, because the capillary transmittance is known to decrease sharply at the onset of pump beam self-focusing [8]. In our case, the capillary transmittance increases with pulse shortening (increasing power). The observed slight decrease in the transmittance with an increase in the pulse duration is related to the energy loss during SRS and rise in the wave attenuation coefficient in the quartz capillary with an increase in the radiation wavelength. Based on the data obtained, further experiments were performed with a pump pulse width of 400 fs, at which one would expect simultaneously a Stokes pulse with a maximally wide spectrum and a high conversion efficiency.

To choose the optimal conversion regime, we measured the dependence of the conversion efficiency in a delayed pulse on the hydrogen pressure (Fig. 3). The conversion effi-

ciency reaches a maximum (~10%) at p_H = 27–30 atm. At these pressures, the energy of the double pump pulse with equal energies of single pulses is 320 μJ and the PRR is 10 kHz. The reason for the decrease in the conversion efficiency at hydrogen pressures higher than optimal is as follows: with an increase in the hydrogen pressure and, correspondingly, the gain increment, the maximum of conversion efficiency shifts to the capillary input, as a result of which the Stokes radiation is attenuated when propagating through the capillary.

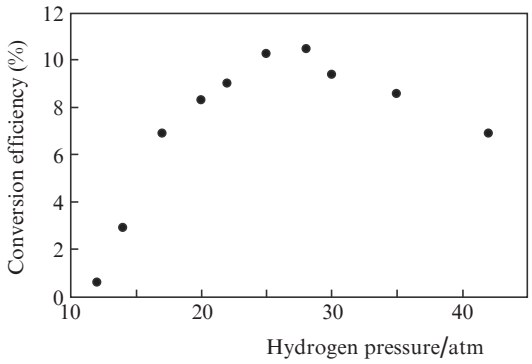

Figure 3. Dependence of the conversion efficiency into a delayed Stokes pulse on the hydrogen pressure. The double pump pulse energy is 320 μJ, and the PRR is 10 kHz.

To obtain a single Stokes pulse with a maximum energy, we investigated the dependence of the energy E_{st} of a Stokes pulse with a wavelength of 1.8 μm on the pump pulse energy E_p. The hydrogen pressure was 27 atm, the PRR was 10 kHz, and the chirped pulse duration was 400 fs. Experiments were carried out with single (the calcite crystal was removed from the optical scheme in this case) and double pump pulses. For the single pulse, the maximum energy was E_{st} = 12 μJ at $E_p \approx$ 200 μJ. With an increase in E_p, the E_{st} value decreased, because the pump pulse power approached the critical self-focusing power. The use of a double pump pulse makes it possible to increase significantly the energy E_{st}. This is related to the rise in the pump pulse energy and, correspondingly, the Raman gain increment, with the pulse power retained below the critical self-focusing power. Figure 4 shows the dependence of the energy of a delayed Stokes pulse on the double pump pulse energy. The fraction of the energy present in the first Stokes pulse did not exceed 10% of the energy of delayed Stokes pulse in the entire range of pump pulse energies under consideration. The maximum obtained energy was E_{st} = 40 μJ at the energy E_p = 390 μJ, which corresponds to a conversion efficiency higher than 10%.

An important characteristic of nonlinear laser converters is the maximum average power of converted radiation, which affects significantly the converter application range. The measured dependence of the conversion efficiency on the ytterbium laser PRR at a fixed (maximally possible for this laser) pulse energy is presented in Fig. 5. The hydrogen pressure is 27 atm, and the energy of the double laser pulse at the capillary input is 320 μJ. It can be seen that the conversion efficiency drops with an increase in the PRR and, correspondingly, the average laser power. The maximum conversion

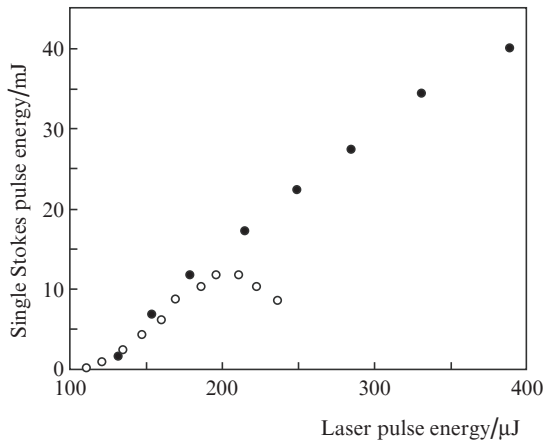

Figure 4. Dependences of the energy of a single (delayed) Stokes pulse on the pump pulse energy under (○) single- and (●) and double-pulse pumping.

efficiency (14%) was obtained at a PRR of 1 kHz (smaller PRR values could not be implemented in the laser system we used). The conversion efficiency decreased to 9.5% with an increase in PRR to 20 kHz. As a result, an average Stokes power of 0.6 W was implemented at an average pump power of 6.3 W. We relate the decrease in the conversion efficiency with a rise in PRR to the gas heating in the capillary. At an average pump power of 6 W and conversion efficiency of 9% in the capillary we used (with an internal volume of 4×10^{-2} cm^3), the specific power of energy release due to the Stokes losses is 10 W cm^{-3}. Heating of the high-pressure hydrogen, which occurs mainly near the capillary axis, where the pump beam intensity is maximum, may lead to inhomogeneous distribution of the gas density over the capillary cross section, which, in turn, should cause a nonuniform distribution of refractive index over the capillary cross section and, as a consequence, lead to excitation of higher order modes with a large attenuation coefficient for the pump radiation in the capillary. As a result, the Raman gain increment and conversion efficiency decrease. This suggestion is confirmed by the measured dependence of the capillary transmittance on PRR (see Fig. 5): the transmittance and conversion efficiency decrease simultaneously with an increase in the pump PRR,

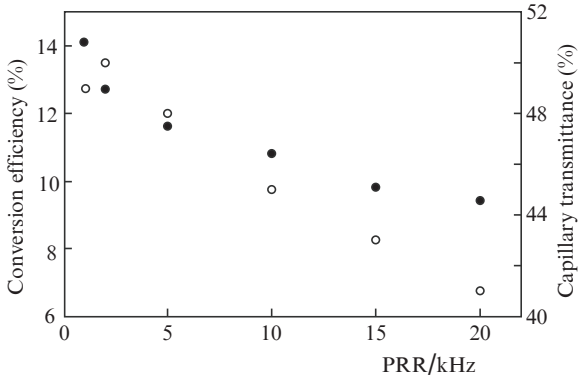

Figure 5. Dependences of the (●) conversion efficiency and (○) capillary transmittance on the pump PRR.

despite the fact that the capillary transmittance should rise with a decrease in the SRS efficiency during Raman scattering in the absence of any additional (except for quantum) losses. For example, this pattern was observed when studying the influence of the pump pulse width on the conversion efficiency (see Fig. 2).

To estimate the Raman converter stability, we measured the relative dispersion distribution over the energy of single Stokes pulses (the ratio of the distribution width to the average pulse energy). The relative dispersion, calculated for a sequence of 10^4 pulses, was found to be 10% in the entire range of parameters under consideration.

4. Stokes pulse spectrum and duration

Figure 6 shows the Stokes component spectra recorded at different pump pulse widths. A double pump pulse with an energy of 320 μJ was used in both cases. The single pulse duration was varied from 400 to 600 fs, and the hydrogen pressure was 27 atm. The Stokes pulse spectrum has a shape characteristic of spectra broadened during nonlinear phase modulation [8]. As was noted above, the spectral width under conditions of nonlinear phase self-modulation is given by the expression $\Delta\nu \propto P/\tau \propto E/\tau^2$. Therefore, with an increase in the pulse duration by a factor of 1.5, the spectrum should narrow by a factor of 2.25. The experimentally observed narrowing is close to the calculated one. Based on these results, one can conclude that the Stokes component arises when the pump radiation (spectrally broadened during self-phase modulation) undergoes SRS.

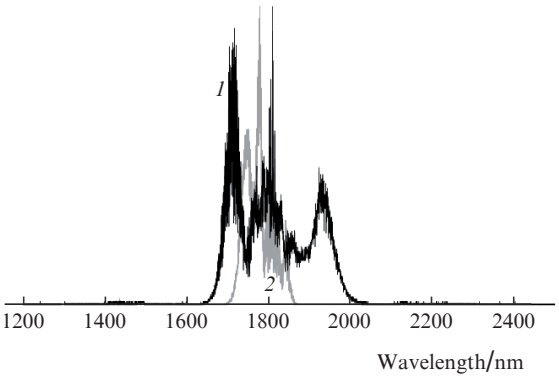

Figure 6. Stokes pulse spectra for laser pulse durations of (*1*) 400 and (*2*) 600 fs.

The positive frequency chirp of the Stokes pulse, which arises under nonlinear phase modulation, can be compensated for (the Stokes pulse is compressed to the transform-limited pulse with the corresponding spectrum) during Stokes pulse propagation in fused silica, which has negative second-order dispersion at the Stokes wavelength.

The negative dispersion optimal for compression was chosen by varying the thickness of the quartz plate mounted at the output of the chamber with capillary (plate P in Fig. 1). The shortest Stokes pulses were obtained at a total optical path length of 3 cm in the quartz elements at the converter chamber output (chamber, lens, broadband filter, Rochon prism, quartz plate). In this case, the Stokes pulse duration varied within 25–30 fs for the PRR ranging from 1 to 20 kHz.

The autocorrelation function (ACF) of a compressed Stokes pulse is shown in Fig. 7. As the calculation showed, to obtain a 27-fs pulse under compression in a 3-cm-thick quartz plate, the duration of the initial chirped pulse should be ~200 fs; i.e., the Stokes pulse at the capillary output should be approximately twice as short as the pump pulse, which is typical of SRS processes. The ACF shape suggests that the compressed pulse includes the central peak and a longer subpulse, containing ~30% pulse energy. The occurrence of the subpulse is related to the frequency chirp nonlinearity and insufficient chirp compensation in fused silica. With allowance for the ACF shape, the peak power of the Stokes pulse reached 1 GW after the compression.

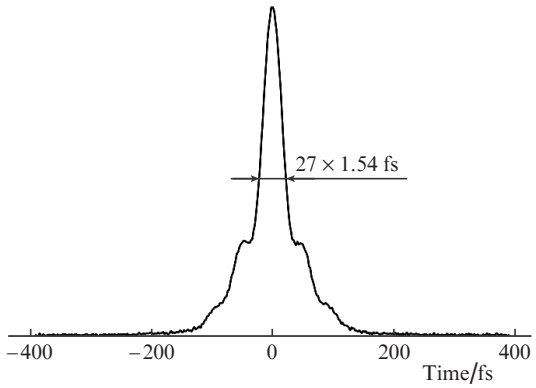

Figure 7. Autocorrelation function of a 27-fs Stokes pulse (the pulse shape is assumed to be described by sech^2). The PRR is 10 kHz, and the Stokes pulse energy is 40 μJ.

5. Conclusions

The main results of this study are as follows.

(1) A scheme of double-pulse pumping of a Raman-active medium was developed. It was shown that, under the conditions of SRS of femtosecond ytterbium laser radiation in hydrogen, the conversion efficiency is maximum for pulses longer than 400 fs.

(2) The conversion efficiency into Stokes radiation with a centre wavelength of 1.8 μm was found to be 14% and 9.5% at pulse repetition rates of 1 kHz and 20 kHz, respectively. The maximum average Stokes power was 0.6 W.

(3) The Stokes pulse was compressed to a duration less than 30 fs in fused-silica optical elements at the output of Raman-active medium. The peak radiation power reached 1 GW.

Note also that this Raman converter scheme appears promising for other types of lasers, in particular, for a Cr:forsterite femtosecond laser [9] with a radiation wavelength of 1.24 μm. In this case, the Stokes component wavelength is 2.6 μm. As was shown in [1], nonlinear phase modulation should not reduce significantly the conversion efficiency at this pump/Stokes wavelength ratio.

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