

https://doi.org/10.1070/QEL17508

SRS of ultrashort laser pulses with nonlinear phase modulation

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

Abstract. **The SRS process in the regime of nonlinear phase modulation of laser and Stokes radiation waves is studied experimentally. The effect of the relationship between the laser and Stokes wavelengths on the value of the minimum laser pulse duration, at which an efficient SRS process is possible, is ascertained. The possibility of obtaining maximally broadband Stokes radiation pulses by matching the group velocities of the laser and Stokes pulses is demonstrated.**

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

1. Introduction

The main competing processes that reduce the efficiency of SRS conversion of laser pulses with a duration of less than 10 ps in gaseous media are multiphoton photoionisation of the active medium molecules, self-focusing of pump radiation, and nonlinear phase modulation, which broadens the pump and Stokes radiation spectra. To exclude photoionisation, the efficiency of which is determined by the pump intensity, long-focus optics can be used, which makes it possible to increase the transverse size of the interaction region and, therefore, to reduce the laser radiation intensity. The critical self-focusing power, which is inversely proportional to the gas pressure, can be made higher than the pump power by choosing the pressure of the active medium. The broadening of the spectrum of a laser pulse due to nonlinear self-phase modulation, which is equal to the ratio of the width of the spectrum of the radiation pulse at the exit from the medium to the width of the spectrum at the entrance to it, is determined by the expression [1]:

$$B \approx \frac{2\pi n_2 EL}{\lambda \tau S} \text{ (for } B > 1),$$

where n_2 is the nonlinear part of the refractive index; λ is the radiation wavelength; E is the pulse energy; τ is the pulse duration; S is the cross sectional area of the light beam; and L is the interaction length. In turn, the Raman increment M at a pump pulse duration shorter than the dephasing time T_2 of the active medium coherent oscillations (for transient SRS) is described by the expression [2]

$$M = \frac{gEL}{T_2 S},$$

where g is the steady-state Raman gain. Since the refractive index n_2 is proportional to the gas pressure and the time T_2 is inversely proportional to it, the only way to suppress the nonlinear self-modulation process while maintaining the Raman increment is to increase the laser pulse duration. For this purpose, a conversion technique was developed, which consists in lengthening the pump pulse by frequency chirping, SRS of the chirped pulse, and subsequent time compression of the Stokes pulse [3, 4]. This technique was used to obtain a Stokes pulse with a duration close to the duration of the initial laser pulse [5].

To expand the field of application of Raman converters of ultrashort pulses, the SRS process, which occurs simultaneously with the process of nonlinear phase modulation, seems to be promising. In this case, it becomes possible to generate broadband Stokes pulses, the duration of which after time compression will be significantly shorter than the duration of the pump pulse. To generate broadband Stokes radiation, a pump pulse spectrally broadened in the course of self-phase modulation is required. Since the broadening of the pulse spectrum during self-phase modulation is proportional to the radiation intensity, to obtain a wider spectrum at a fixed pulse energy, it is necessary to reduce its duration. However, as shown in [6], when the pump pulse duration is less than a certain value, the efficiency of the SRS process drops sharply, despite the fact that the radiation intensity is below the multiphoton photoionisation threshold and the radiation pulse power is less than the critical self-focusing power. In our previous papers [7, 8], we proposed an interpretation of this effect based on the influence of nonlinear self-modulation of the pump and Stokes wave radiation on the SRS process. It is as follows. During the propagation of high-power pulses in the active medium, nonlinear additions appear to the magnitudes of the wave vectors of the pump radiation k_p and the Stokes component k_s [8]:

$$k_p^{NL}(t) = \frac{2\pi n_2 I_p(t)}{\lambda_p}, \quad k_s^{NL}(t) = \frac{4\pi n_2 I_p(t)}{\lambda_s},$$

where $I_p(t)$ is the pump intensity; and λ_p and λ_s are the pump and Stokes wavelengths. The nonlinear addition to the pump wave vector is due to the process of nonlinear self-phase modulation, and the addition to the Stokes wave vector is due to the cross-modulation process, since when the Stokes wave

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Received 16 December 2020
Kvantovaya Elektronika **51** (3) 217–221 (2021)
Translated by V.L. Derbov

develops from the level of spontaneous scattering, its intensity is much lower than that of pump radiation. The difference in the additions give rise to a wave mismatch of the pump and Stokes radiation interacting at different times. This causes a decrease in the Raman increment in the case of transient SRS and a failure in the Stokes wave generation. Based on this assumption, an expression was obtained for the minimum possible duration of the pump pulse, which depends on the parameters of the gas medium and laser radiation as follows [8]:

$$\tau_{\min} \approx \frac{160 T_2 n_2}{g} \left| \frac{2}{\lambda_s} - \frac{1}{\lambda_p} \right|. \quad (1)$$

When deriving this expression, we did not take into account the effects associated with the dispersion of the active medium, such as a change in the pulse shape and the appearance of a time shift between the pump pulse and the Stokes one. Since the maximum value of the Raman gain in the region of collisional broadening of the spontaneous Raman scattering line does not depend on the gas pressure, the minimum pump pulse duration is also independent of it. Therefore, the minimum duration of the pump pulse will not change with a change in its energy and a change in the gas pressure and, hence, the Raman increment to achieve the maximum conversion efficiency. In addition, due to the dependence of the Raman gain on the radiation wavelength, $g \propto \lambda_s^{-1}$ [2], the minimum pulse duration depends on the radiation wavelength as

$$\tau_{\min} \propto \left| 2 - \frac{\lambda_s}{\lambda_p} \right|. \quad (2)$$

Hence, it follows that the effect of nonlinear phase modulation on the Raman amplification process weakens, and the minimum pump pulse duration decreases as the condition $\lambda_s = 2\lambda_p$ is approached. In this case, the nonlinear parts of the wave vectors of the pump radiation and the Stokes wave become equal, and the wave mismatch disappears.

Preliminary experiments on SRS with nonlinear phase modulation were performed using pump radiation at a wavelength of 1.03 μm. An ytterbium laser pulse with a duration of 250 fs was converted by SRS in hydrogen with an efficiency of ~10% into a Stokes component pulse with a wavelength of 1.8 μm and a duration of 30 fs [9, 10]. A detailed study of the dependence of the minimum possible pump pulse duration on the parameters of laser radiation and active medium has not been carried out. Such studies are part of this work.

It is known [11] that for SRS of a broadband Stokes signal in a broadband pumping field, in particular for SRS under conditions of nonlinear phase modulation, the conversion efficiency depends on the difference between the group velocities of the pump and the Stokes pulses. Using waveguide structures makes it possible to ensure the equality of group velocities and thus increase the Raman increment and obtain the widest possible broadband Stokes pulses. The results of these studies are also presented below.

2. Experimental results and discussion

The setup for experimental studying SRS with nonlinear phase modulation is schematically shown in Fig. 1. A femtosecond Ti:sapphire laser system (REUS-3m1k, Avesta) with the centre wavelength of 0.8 μm and an ytterbium laser system with a radiation wavelength of 1.03 μm (TETA-3-HE, Avesta) served as pump radiation sources. Both laser systems were constructed according to the generator–regenerative amplifier scheme. The pulse repetition rate was 1 kHz, and the energy of a single pulse reached 2 mJ in both systems. The duration of the output radiation pulse could be varied by changing the distance between the gratings of the time compressor at the output of the system. The minimum pulse duration was 30 and 250 fs for the Ti:sapphire laser and the ytterbium laser, respectively.

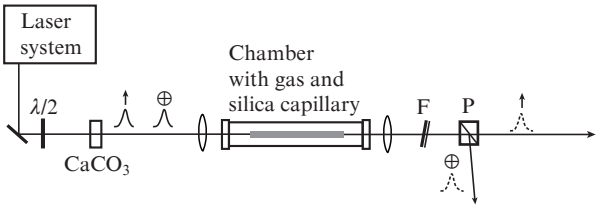

Figure 1. Optical scheme of the setup:
($\lambda/2$) half-wave phase plate; (F) broadband filter for separating Stokes radiation; (P) prism polariser; the icons above the pulses indicate the direction of polarisation.

The experiments were carried out using a quartz capillary placed in a gas-filled chamber 120 cm long. The capillary length was 80 cm, and the inner diameter was 250 μm. The active substance was gaseous hydrogen.

In front of the entrance to the chamber, a birefringent calcite plate 1 cm thick and a half-wave phase plate were installed. This system allowed implementing both a single-pulse pumping regime and a regime with two time-separated orthogonally polarised pulses, which allows wider-band pulses to be obtained as compared to single-pulse pumping [10]. A collimating lens, a broadband filter for separating Stokes radiation, and a prismatic polariser for selecting a pulse with the required polarisation were placed at the exit of the hydrogen chamber. The pulse spectra were recorded using an ASP-IR-2.6 scanning spectrometer (Avesta).

The experiments were carried out with a power of laser pulses lower than the critical self-focusing power. For radiation with wavelengths of 0.8 and 1.03 μm, the critical self-focusing powers (in GW) in hydrogen are $P_{cr}^{0.8} \approx 12/H$ and $P_{cr}^{1.03} \approx 19/H$, where H is the hydrogen pressure in atmospheres [12]. The onset of self-focusing upon varying the radiation power was detected by a sharp decrease in the capillary transmission.

The dependences of the conversion efficiency on the pulse duration in the single-pulse regime are shown in Fig. 2. When pumped at a wavelength of 0.8 mm, the conversion efficiency reaches saturation for pulses longer than 2 ps. A decrease in efficiency is observed for shorter pulses. The hydrogen pressure in the capillary was 15 atm. The Stokes radiation wavelength was 1.2 μm, and the radiation pulse energy was 0.5 mJ. The critical self-focusing power has not been exceeded under these conditions.

When proceeding to pump radiation with a longer wavelength (1.03 μm), the conversion efficiency saturates at pulse durations exceeding 0.8 ps. The Stokes radiation wavelength was 1.8 μm. The dependences were measured at hydrogen pressures of 34 and 9 atm, while the pump pulse energies

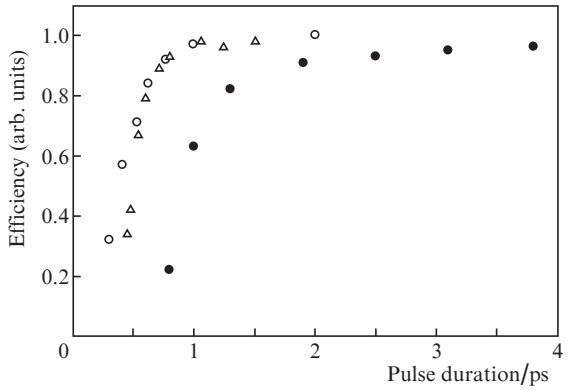

Figure 2. Dependences of the SRS conversion efficiency on the pulse duration when pumped by radiation with wavelengths of 1.03 μm [hydrogen pressure 9 (○) and 34 atm (△)] and 0.8 μm (hydrogen pressure 15 atm (●)].

were 0.3 and 1.2 mJ, respectively. Figure 2 shows that the dependences coincide in both cases. This confirms that the minimum pump pulse duration is independent of the gas pressure.

It should be noted that a similar dependence of the conversion efficiency in hydrogen on the duration of a radiation pulse with a wavelength of 1 μm was observed in [13], where the SRS process in a microstructured fibre was investigated at a pulse energy of ~10 μJ. The conversion efficiency saturated at pulse durations longer than 0.7 ps.

The ratio of the obtained values of the minimum possible pump pulse durations for transient SRS is in good agreement with the ratio of the values calculated using Eqn (2), from which it follows that in hydrogen the minimum pump pulse duration decreases by about 2.5 times. Estimation of the minimum possible pump pulse duration using Eqn (1) for pumping with a wavelength of 1.03 μm yields a value of ~0.5 ps, which agrees with the experimentally measured value.

To obtain broadband Stokes pulses and, consequently, the shortest pulses after time compression, it is necessary to implement the SRS regime, in which effective conversion is carried out throughout the entire pump pulse chirped by self-modulation. This can be achieved by using a double-pulse pumping scheme and matching the group velocities of the pump and Stokes radiation pulses. In the previously developed method of pumping by two successive orthogonally polarised pulses [4], the first pulse produces a coherent wave of molecular vibrations in the active medium during stimulated Raman scattering, on which the delayed pump pulse is efficiently scattered over its entire length. Equalisation of the group velocities of pump and Stokes pulses can be achieved in gas-filled waveguides.

Using the expression for the magnitude of the fundamental mode wave vector in a gas-filled hollow optical waveguide [14]

$$k = \frac{2\pi}{\lambda}\left[n - \frac{1}{2}\left(\frac{2.4\lambda}{\pi d}\right)^2\right]$$

(where n is the gas refractive index, and d is the inner diameter of the waveguide), and for the refractive index of a gas at a pressure of one atmosphere, the expression [15]

$$n - 1 = A + \frac{D}{\lambda^2}$$

(where A and D are constants for a particular gas), we can calculate the difference between the group velocities of the pump pulse and the Stokes pulse and obtain the pressure at which the group velocities equalise:

$$H_v \approx \frac{0.1\lambda_s^2\lambda_p^2}{Dd^2}.$$

In SRS of radiation with a wavelength of 1.03 μm in hydrogen filling a capillary with an inner diameter of 250 μm, the group velocities equalise at a hydrogen pressure of 5.2 atm (for hydrogen, $D = 10^{-14}$ cm^2 [15]).

Figure 3 shows the spectra of pump and Stokes radiation pulses at the exit from the capillary. In this case, to achieve the maximum width of the Stokes radiation spectrum, we used the two-pulse pumping regime with equal energies of the pulses [10]. The durations of single pump pulses were 400 fs. The experiments were carried out at gas pressures of 9 and 34 atm. In this case, the energies of double pump pulses were 1200 and 300 mJ, respectively. At the given pump pulse energies and gas pressures, SRS conversion of the delayed pump pulse is observed with an efficiency of 9%

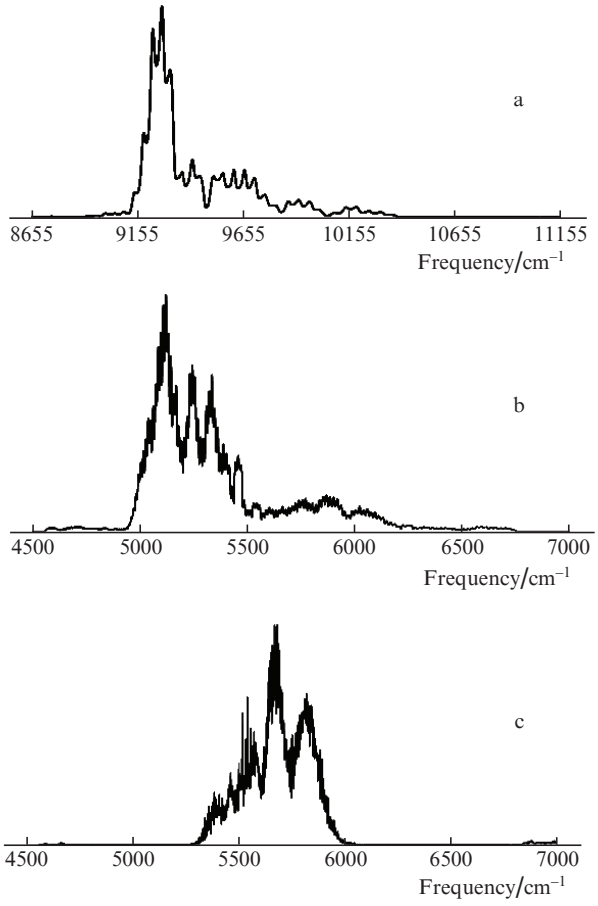

Figure 3. Spectra of (a) pump and (b, c) Stokes radiation pulses at hydrogen pressures of (b) 9 and (c) 34 atm at the exit from the capillary (for clarity, the horizontal axes are shifted by the Stokes shift in hydrogen).

at a pressure of 34 atm and 11 % at a pressure of 9 atm. For the first pump pulse, the conversion efficiency did not exceed 1 %. The spectrum of the first pump pulse broadened by self-phase modulation is shown in Fig. 3a. The pump pulse spectra have similar shapes in the two conversion regimes investigated, since the broadening B of the spectra coincides in both cases. There is a noticeable difference in the spectra of Stokes radiation. This feature in the formation of the Stokes pulse spectrum can be associated with the difference in the group velocities of the pulses. At a pressure of 9 atm, close to the pressure at which the group velocities coincide, the calculated time delay between the pump pulse and the Stokes pulse as they pass through an 80 cm capillary is 20 fs, which is much shorter than the pulse duration. The width and shape of the spectrum of the Stokes pulse (Fig. 3b) are close to those for the spectrum of the pump radiation pulse. At a pressure of 34 atm, the time delay increases to 160 fs. The pulses propagate in the normal dispersion regime. The Stokes pulse leads the pump pulse, and the most efficient conversion occurs at the trailing edge of the positively chirped Stokes pulse, in the high frequency region. As a result, the spectrum of the Stokes pulse narrows and shifts to the short-wavelength region (Fig. 3c).

Time compression of positively chirped Stokes pulses can be implemented by their propagation in materials with anomalous dispersion in the 1.8 μm region, e.g., in fused silica [9, 11]. Figure 4 shows the calculated shapes of transform-limited pulses (extremely short pulses after time compression), corresponding to the experimental spectra of Stokes pulses. A pulse with a duration of 18 fs can be obtained by compressing a wider-band Stokes pulse in the case of SRS in the regime of matching the group velocities of the pump pulses and the Stokes component at a pressure of 9 atm. At a hydrogen pressure of 5 atm, which corresponds to an exact matching of group velocities, the SRS threshold was not exceeded under the conditions of our experiment at a maximum possible pump pulse energy of 2 mJ. At higher hydrogen pressure and an increase in the group velocity difference, the spectrum of the Stokes component narrows, and the calculated duration of the transform-limited Stokes pulse increases to 25 fs.

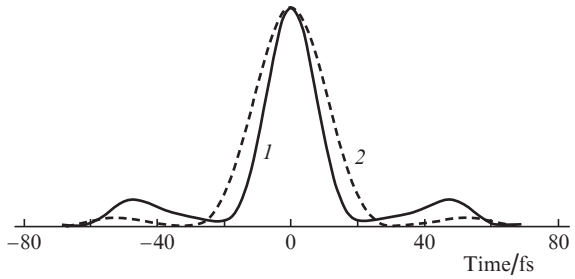

Figure 4. Calculated shapes of transform-limited Stokes pulses, the spectra of which are shown in Figs 3b and 3c, for hydrogen pressures of (*1*) 9 and (*2*) 34 atm.

3. Conclusions

In this paper, we present the results of experimental studies that confirm the mechanism [8] of the influence of nonlinear phase modulation on the process of transient SRS. It is shown that the minimum possible duration of a pump radiation pulse at a fixed wavelength depends only on the relation between the Raman gain and the nonlinear refractive index of the active medium. In this case, the achievement of the shortest pump pulse duration is possible when the Stokes radiation wavelength is twice the pump radiation wavelength. Experimentally, for stimulated Raman scattering in hydrogen, the minimum pump pulse duration was ∼0.8 ps for pump wavelengths of 1.03 μm and a Stokes component of 1.8 μm and ∼2 ps for wavelengths of 0.8 and 1.2 μm, respectively. It is also shown that the widest-band chirped Stokes pulses can be obtained in a waveguide by equalising the group velocities of the pump pulses and the Stokes component.

From the experimental data of this work and previous studies [9, 10] of the SRS conversion efficiency and the Stokes radiation spectral width, it seems promising to use the SRS of the radiation from a widely used femtosecond ytterbium laser for generating pulses in the 2 μm region with a duration of ∼20 fs and an energy of ∼100 μJ. Light pulses with a duration of a few light wave cycles and a power of ∼5 GW can be used in mid-IR nonlinear optics experiments. Also, based on the dependence (2) of the minimum pump pulse duration on the wavelength, we can assume that the shortest Stokes pulses of mid-IR radiation with a wavelength of 2.6 μm can be obtained by SRS of radiation from a Cr:forsterite laser with a wavelength of 1.25 μm in high-pressure hydrogen. The operation stability of such Raman converters under optimal conditions can approach the stability of a pump laser [16].

Note also the possibility to implement the Raman conversion scheme for femtosecond laser pulses in microstructured hollow fibres, in particular, in hollow revolver-type fibres [17]. The low attenuation coefficient allows the use of fibres with a length of several meters for conversion. In these fibres, by choosing the gas pressure, it is possible to realise the regime of equal group velocities of the pump and Stokes pulses. Then, in the input part of the fibre, the spectrum of the pump radiation pulse will broaden and the pulse duration will increase due to its dispersive spreading. With further pulse propagation, when its duration exceeds the minimum possible pulse duration, the process of SRS of broadband pump radiation will begin. Matching the group velocities will allow efficient conversion into broadband Stokes radiation.

References

1. Pinault S.C., Potasek M.J. *J. Opt. Soc. Am. B*, **2**, 1318 (1985).
2. Zel'dovich B.Ya., Pilipetskiy N.F., Shkunov V.V. *Principles of Phase Conjugation* (Berlin, New York: Springer-Verlag, 1985; Moscow: Nauka, 1985)].
3. Jordan C., Stankov K.A., Marowsky G., Canto-Said E.J. *Appl. Phys. B*, **59**, 471 (1994).
4. Konyashchenko A.V., Losev L.L., Tenyakov S.Yu. *Opt. Express*, **15**, 11855 (2007).
5. Didenko N.V., Konyashchenko A.V., Kostryukov P.V., Losev L.L., Pazyuk V.S., Tenyakov S.Yu., Molchanov V.Ya., Chizhikov S.I., Yushkov K.B. *Quantum Electron.*, **45**, 1101 (2015) [*Kvantovaya Elektron.*, **45**, 1101 (2015)].
6. Losev L.L., Song J., Xia J.F., Strickland D., Brukhanov V.V. *Opt. Lett.*, **27**, 2100 (2002).
7. Konyashchenko A.V., Kostryukov P.V., Losev L.L., Pazyuk V.S. *Quantum Electron.*, **47**, 1 (2017) [*Kvantovaya Elektron.*, **47**, 1 (2017)].

8. Konyashchenko A.V., Kostryukov P.V., Losev L.L., Pazyuk V.S. *Quantum Electron*., **47**, 593 (2017) [*Kvantovaya Elektron*., **47**, 593 (2017)].
9. Konyashchenko A.V., Losev L.L., Pazyuk V.S. *Opt. Lett*., **44**, 1646 (2019).
10. Konyashchenko A.V., Losev L.L., Pazyuk V.S. *Quantum Electron*., **49**, 845 (2019) [*Kvantovaya Elektron*., **49**, 845 (2019)].
11. Beldyugin I.M., Zubarev I.G., Mikhailov S.M. *Sov. J. Quantum Electron.*, **7**, 1471 (1980) [*Kvantovaya Elektron*., **10**, 847 (1980)].
12. Lehmeier H.J., Leupacher W., Penzkofer A. *Opt. Commun*., **56**, 67 (1985).
13. Loranger S., Russell P.S.J., Novoa D. *J. Opt. Soc. Am. B*, **37**, 3550 (2020).
14. Im S., Husakou A., Herrmann J. *Opt. Express*, **17**, 13050 (2009).
15. Born M., Wolf E. *Principles of Optics* (Oxford: Pergamon, 1969; Moscow: Nauka, 1973).
16. Didenko N.V., Konyashchenko A.V., Losev L.L. *Quantum Electron*., **50**, 834 (2020) [*Kvantovaya Elektron*., **50**, 834 (2020)].
17. Gladyshev A., Yatsenko Y., Kolyadin A., Kompanets V., Bufetov I. *Opt. Mater. Express*, **10**, 3081 (2020).