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Minimum possible laser pulse duration for SRS

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Abstract. **The minimum laser pulse duration for transient stimulated Raman scattering is shown to be limited as a consequence of nonlinear phase modulation of the laser and Stokes waves. It is determined by the ratio of the nonlinear refractive index to the spontaneous Raman scattering cross section.**

Keywords: *stimulated Raman scattering, femtosecond laser, self-phase modulation, cross-phase modulation.*

1. Introduction

It is well known that the energy efficiency of laser light conversion in the stimulated Raman scattering (SRS) process drops sharply in the case of pulses with durations of hundreds of femtoseconds [1 – 3]. In view of this, unique schemes have been proposed for SRS converters of femtosecond laser pulses, which first increase the duration of an input femtosecond laser pulse to a few picoseconds by introducing a frequency chirp and then ensure SRS conversion of the chirped pulse and temporal compression of the Stokes pulse [4, 5].

It is possible to identify the key physical processes that can, in principle, cause a reduction in conversion efficiency, to the extent of complete suppression of the SRS of subpicosecond laser pulses: nonlinear photoionisation of the active medium, self-focusing of light and nonlinear phase modulation. (Effects related to dispersion in the active medium can be left out of account in the case of gaseous active media and pulses with durations of ~100 fs.) The first two processes can be precluded by using long focal length optics and optical fibres capable of reducing the light intensity in the conversion region and ensuring conditions under which the SRS threshold power is lower than the critical power for self-focusing. Nonlinear phase modulation cannot be completely eliminated because, like the Raman gain per unit length, it depends on the laser light intensity and the length of the active medium [6]. It is also worth noting that Losev et al. [3] experimentally observed a sharp decrease in Stokes conversion efficiency in the case of pump spectrum broadening at the output of the active medium. Thus, there is evidence that it is the phase modulation process and the associated pump spectrum broadening that are responsible for the decrease in conversion efficiency. In a previous study [7], we qualitatively described the effect of nonlinear phase modulation on the SRS process.

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The objectives of this work were to describe in greater detail the mechanism of SRS suppression in the case of nonlinear phase modulation of pump light, estimate the minimum possible laser pulse duration in the SRS process and experimentally verify theoretical predictions.

2. Estimation of the minimum pump pulse duration for SRS

Consider the generation of a Stokes wave under pumping from the spontaneous scattering level at a pump power slightly above the SRS threshold. In such a case, the relationship $I_p(z) \gg I_S(z)$, where $I_p(z)$ and $I_S(z)$ are the intensities of the pump and Stokes waves, respectively, is fulfilled over the entire length L of the active medium.

The magnitude of the pump wave vector k_p can be represented as the sum of a linear and a nonlinear component: $k_p(z) = k_p^0 + k_p^{NL} = 2\pi n_p/\lambda_p + 2\pi n_2 I_p(z)/\lambda_p$, where λ_p is the laser wavelength and n_p and n_2 are the linear and nonlinear components of the refractive index, respectively. The time variation of the intensity and, hence, wave vector leads to self-phase modulation. The magnitude of the wave vector of the Stokes wave is $k_S(z) = k_S^0 + k_S^{NL} = 2\pi n_S/\lambda_S + 4\pi n_2 I_p(z)/\lambda_S$, where λ_S is the Stokes wavelength and n_S is the linear component of the refractive index of the Stokes wave. The nonlinear component of the wave vector of low-intensity Stokes light is determined by the cross-phase modulation process induced by the intense laser wave [8].

In Raman-active media under pumping by subpicosecond pulses, SRS is a transient process, with the pump pulse duration shorter than the dephasing time T_2 of coherent vibrations of excited molecules in the active medium (phonon wave). It is, therefore, reasonable to assume that, in the case of transient SRS, the wave vector of the phonon wave remains constant during the laser pulse. If the wave vector difference between the Stokes and laser pump waves varies, whereas the wave vector of the previously formed phonon wave remains unchanged, conversion efficiency will drop. Such a situation may occur when the wave vector of interacting waves has an intensity-dependent nonlinear component. There is then a wavenumber mismatch between the waves at low and maximum pump intensities (I_p^{max}):

$$\Delta k(z) = (k_p - k_S)_{I_p \approx I_p^{max}} - (k_p - k_S)_{I_p \approx 0}$$

$$= 2\pi n_2 I_p^{max}(z)\left(\frac{1}{\lambda_p} - \frac{2}{\lambda_S}\right). \quad (1)$$

Conversion efficiency will drop and the SRS threshold will rise sharply at a nonlinear phase shift [9]

$$\Phi = \left| \int_0^L \Delta k(z)\,\mathrm{d}z \right| \geqslant \pi .$$

Taking into account (1) and the fact that, under unsteady-state conditions, SRS develops from the spontaneous scattering level if the condition $g(\tau/T_2)\int_0^L I_p^{\max}(z)\,\mathrm{d}z > 80$ for the Raman increment is met [10] (where g is the steady-state SRS gain coefficient and τ is the pump pulse duration), we find that, for the nonlinear phase shift not to exceed π and for the SRS process to occur, the following condition should be met:

$$\tau \geqslant \frac{160 T_2 n_2}{g}\left(\frac{2}{\lambda_S} - \frac{1}{\lambda_p}\right). \qquad (2)$$

Note that, since $g \sim (\mathrm{d}\sigma/\mathrm{d}\Omega)T_2 N$ [9] (where $\mathrm{d}\sigma/\mathrm{d}\Omega$ is the differential spontaneous Raman scattering cross section and N is the concentration of active particles) and $n_2(N) \approx n_2(1) \times N/N(1)$ [$n_2(1)$ and $N(1)$ are, respectively, the nonlinear refractive index and particle concentration at unit pressure], it follows from (2) that the minimum pump pulse duration is independent of the pressure of the gaseous active medium:

$$\tau_{\min} \sim n_2(1)\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{-1}. \qquad (3)$$

Analysis of Raman-active media with the use of (2) and (3) suggests that the shortest pump pulses, of ~500 fs duration, can be converted using SRS in hydrogen and methane. Both the Raman scattering cross section and nonlinear refractive index of methane are about three times greater [11]. These estimates agree rather well with available experimental data [12]. The duration of the shortest pulses converted under single-frequency pumping was ~300 fs [2, 12]. Under double-frequency pumping, when two femtosecond pulses with comparable amplitudes and a frequency difference equal to the Raman frequency are sent to the input of the active medium, the pump pulse duration can be reduced, because in this case a lower Raman increment is needed in comparison with single-frequency pumping [13].

To ascertain that the nonlinear phase modulation of femtosecond pulses has a critical effect on the SRS process, we carried out experiments in which only the pump laser pulse duration was varied.

3. Experimental results and discussion

In our experiments, we used an Avesta Project REUS-40F1K Ti:sapphire laser system based on chirped-pulse amplification, which included a master oscillator and regenerative amplifier. The laser pulse duration was varied by changing the diffraction grating separation in the temporal compressor at the output of the laser system. In this way, we were able to gradually vary the pulse duration from 0 fs to 4.5 ps. At a pulse repetition rate of 1 kHz, the pulse energy was ~1 mJ. The centre wavelength was 800 nm.

The optical scheme chosen for the Raman converter included a silica capillary tube. We used a capillary 150 μm in inner diameter and 60 cm in length, placed in a 1.2-m-long chamber. A scheme with a gas-filled capillary tube was chosen because it allowed us to lower the SRS energy threshold and ensure conditions under which the pump pulse power was lower than the critical power for self-focusing. In this way, we precluded any effect of self-focusing on the SRS process in our experiments.

The laser beam was focused by a lens with a focal length of 1 m onto the input end of the capillary. The transmission of the capillary was ~30%. The hydrogen pressure in our experiments was 10 atm. The emission spectrum at the output of the hydrogen-filled chamber was measured by an Avesta Project ASP-75 spectrometer. For energy measurements, an ~1.2-μm Stokes pulse was separated using coloured glass.

Figure 1 shows the Stokes pulse energy as a function of negatively chirped pump pulse duration. It is seen that the Stokes pulse energy and, hence, conversion efficiency drop sharply at pump pulse durations under 1 ps, which agrees quite well with the above estimates. At the same time, the measured transmission of the capillary remained constant when the pump pulse duration was varied from 0.5 to 5 ps, indicating that there was no laser beam self-focusing. Note also that a similar decrease in conversion efficiency at pump pulse durations under 1 ps was observed in a previous study [14], even though other experimental conditions were used: pump pulse energy of ~100 μJ and hydrogen pressure of 130 atm.

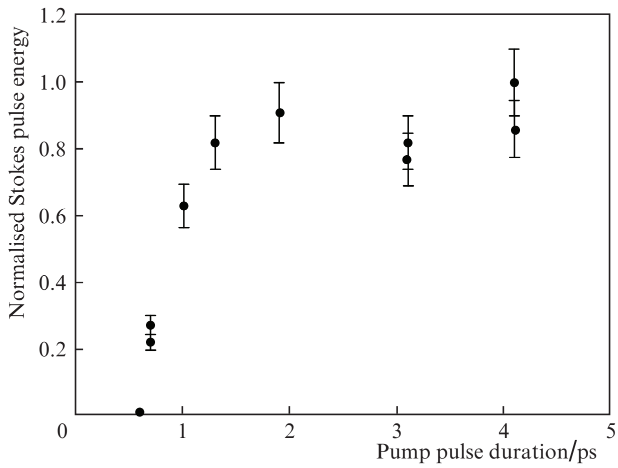

Figure 1. Stokes pulse energy as a function of laser pump pulse duration.

The effect of nonlinear phase modulation of pump pulses on Stokes generation efficiency is also evidenced by the pump spectrum measured at the capillary output (Fig. 2). At a pump pulse duration of ~4 ps, which allows for efficient Stokes generation, there is no pump spectrum broadening (Fig. 2b). The pump spectrum is narrower than the spectrum at the capillary input (Fig. 2a). (The peak near 830 nm is due to the generation of rotational Stokes waves.) The narrowing of the spectrum is due to the conversion of spectral components located at the trailing edge of the frequency-chirped pulse to Stokes emission in a transient SRS process. In the case of negatively chirped pump pulses, only their long-wavelength spectral components are converted. Reducing the pump pulse duration to 0.7 ps leads to a drop in Stokes generation efficiency, and we observe pump spectrum broadening at the capillary output (Fig. 2c). In addition, a narrow peak emerges at the centre wavelength of the pump spectrum, which is character-

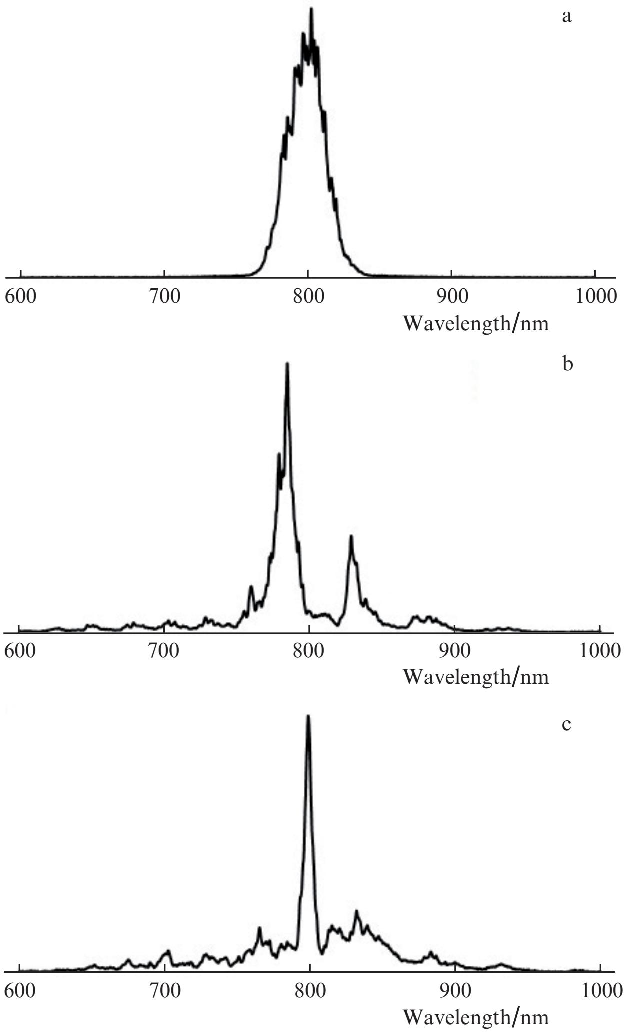

Figure 2. Laser emission spectrum at the capillary input (a) and output at pump pulse durations of 4.5 (b) and 0.7 ps (c).

istic of nonlinear self-phase modulation of negatively frequency-chirped pulses. This effect is known as spectral compression [15].

It follows from relation (1) that, if the condition $\lambda_S \approx 2\lambda_p$ is met, the effect of nonlinear phase modulation on Stokes generation is weaker: the nonlinear wavenumber mismatch tends to zero and Stokes generation is possible at shorter pump pulse durations in comparison with pumping at other wavelengths. To verify this conclusion, we experimentally studied SRS conversion of light from an ytterbium laser with a centre wavelength of 1030 nm. According to (2), in the case of SRS in compressed hydrogen the minimum pump pulse duration can then be reduced by about a factor of 2 compared to the pulse duration under pumping by a Ti:sapphire laser. We used an Avesta Project TETA laser system with a pulse energy of 100 μJ. With this laser system, the pulse duration could not be varied, so our experiments were performed at a constant duration (300 fs) of transform-limited laser pulses and 8-nm emission bandwidth. The SRS conversion scheme was the same as above and hydrogen pressure was 25 atm.

Figure 3 shows the laser and Stokes emission spectra thus obtained. The laser pulse bandwidth at the capillary output exceeded the pump pulse bandwidth at the input of the active medium as a consequence of self-phase modulation. Nevertheless, we observed Stokes generation under such conditions, which was impossible under pumping with Ti:sapphire laser pulses of the same duration.

Thus, the present theoretical analysis and experimental data demonstrate that the minimum possible pump pulse duration at which transient stimulated Raman scattering is possible is determined by nonlinear phase modulation of the laser and Stokes waves. Moreover, the minimum pump pulse duration depends on the ratio of the nonlinear refractive index of the active medium to the spontaneous Raman scattering cross section.

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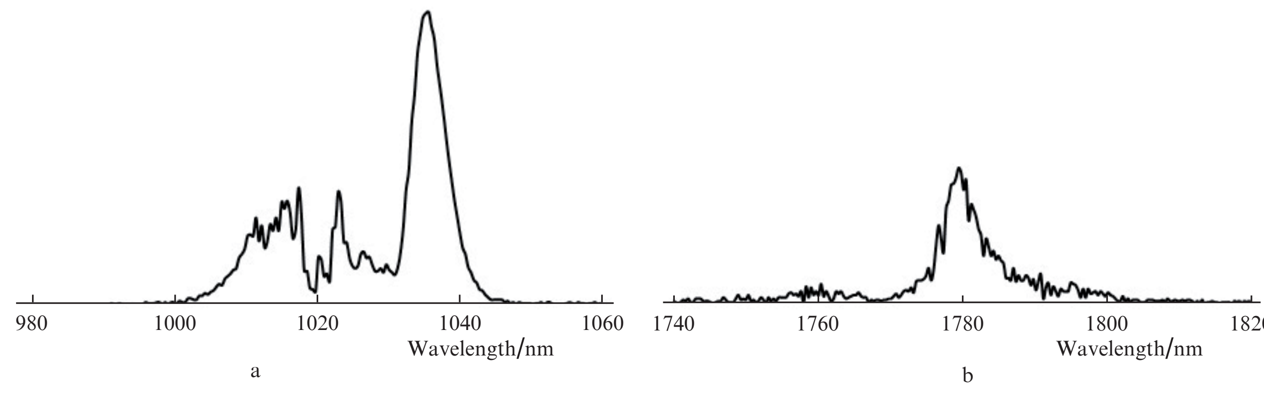

Figure 3. (a) Laser and (b) Stokes emission spectra at the capillary output under ytterbium laser pumping.

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