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Femtosecond compressed-nitrogen Raman laser

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Abstract. **We have estimated the minimum laser pulse duration at which stimulated Raman scattering in gases is possible. Femtosecond Ti:sapphire laser pulses have been converted to the first Stokes in compressed nitrogen using double-pulse pumping of a gas-filled capillary tube by orthogonally polarised chirped pulses. We have obtained 980-nm Stokes pulses of 51 fs duration. The energy conversion efficiency was 12% at a pulse repetition rate of 1 kHz and average laser output power of 2 W.**

Keywords: *stimulated Raman scattering, femtosecond laser, capillary tube, pulse repetition rate, nitrogen gas.*

1. Introduction

Compressed hydrogen and methane are the most widely used gaseous Raman media for frequency conversion of nano- and picosecond laser pulses, because these media have the largest Raman gain coefficients. Active media of femtosecond lasers should meet other requirements. Since conversion is here essentially time-dependent, a key role is played in this case not by the Raman gain coefficient but by the spontaneous Raman scattering cross section [1]. The spontaneous Raman scattering cross sections of most Raman-active gases differ by less than one order of magnitude [2], in contrast to their Raman gain coefficients, which may differ by up to two or more orders of magnitude [3]. Because of this, the list of Raman-active gases suitable for femtosecond laser pulse converters can be extended in comparison with nanosecond pulse converters. In particular, nitrogen gas, which is essentially not used in nanosecond Raman lasers because of its small gain coefficient and the competition between stimulated Brillouin scattering and stimulated Raman scattering (SRS) processes, is of interest for extending the frequency tuning range of femtosecond Raman converters. Nitrogen gas has also attracted considerable attention owing to the possibility of employing it in high-average power Raman converters. The reason for this is that there are no nitrogen molecule dissociation products. Such products may be formed during the generation process and absorb the light.

The purpose of this work was to examine the feasibility of producing a frequency converter for a femtosecond nitrogen Raman laser. Conversion was carried out through the excitation of an active medium by a sequence of two orthogonally polarised frequency-chirped laser pulses [4]. This configuration offers the possibility of obtaining, after compression, Stokes pulses similar in duration to a transform-limited input laser pulse [5]. First, we evaluated the minimum possible chirped excitation pulse duration.

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2. Minimum possible pump laser pulse duration for SRS

Reducing the laser pulse duration and raising the pulse power leads to a number of nonlinear effects, which may prevail over the SRS process. Note, first of all, multiphoton photoionisation, optical breakdown, self-focusing and self-phase modulation. The photoionisation and optical breakdown effects can be eliminated by using long focal length optics, whereas, to reduce the influence of self-focusing and self-phase modulation, one should increase the laser pulse duration via a frequency chirp. Let us estimate the minimum possible chirped pulse duration.

Consider the generation of a Stokes wave at a focal waist of radius ρ in the course of Gaussian laser beam focusing. The laser (pump) light intensity I_p at the focal waist is taken to be constant and determined by the laser pulse power P: $I_p = P/(\pi\rho^2)$. The Stokes intensity I_S, which increases from the spontaneous scattering level, is far lower than the pump intensity. The magnitude of the pump wave vector, k_p, can then be represented as the sum of a linear (k_p^0) and a nonlinear (k_p^{nl}) component:

$$k_p = k_p^0 + k_p^{nl} = \frac{2\pi n_0}{\lambda_p} + \frac{2\pi n_2 I_p}{\lambda_p},$$

where λ_p is the laser wavelength and n_0 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

$$k_S = k_S^0 + k_S^{nl} = \frac{2\pi n_0}{\lambda_S} + \frac{4\pi n_2 I_p}{\lambda_S},$$

where λ_S is the Stokes wavelength. 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 [6].

The SRS of a chirped picosecond laser pulse in an active gaseous medium is a transient process in which the pulse duration is comparable to or shorter than the dephasing time T_2 of coherent vibrations of excited molecules in the active

medium (phonon wave). Therefore, in the case of transient SRS, the phonon wave phase can be taken to remain constant during the laser pulse. The direction of the energy flow between the pump and Stokes waves is determined by the relationship between the phases of the light and phonon waves. If the phase difference between the Stokes and pump waves varies, the Stokes energy may be converted back to laser beam energy, with a reduction in phonon wave amplitude and, hence, in conversion efficiency. Such a situation may occur when the wave vector of interacting waves has an intensity-dependent nonlinear component. There is then a wavenumber mismatch Δk between low and maximum pump intensities:

$$\Delta k = (k_\mathrm{p} - k_\mathrm{S})|_{I_\mathrm{p} \approx I_\mathrm{p}^{\max}} - (k_\mathrm{p} - k_\mathrm{S})|_{I_\mathrm{p} \approx 0} = 2\pi n_2 I_\mathrm{p}^{\max}\left(\frac{1}{\lambda_\mathrm{p}} - \frac{2}{\lambda_\mathrm{S}}\right). \quad (1)$$

The direction of the conversion process reverses and, hence, the Stokes intensity stops rising and the SRS threshold increases sharply when the condition $\Delta k L = \pi$ (where L is the interaction length) is satisfied. Substituting Δk from (1) and taking $I_\mathrm{p}^{\max} = P_{\max}/(\pi\rho^2)$ and $L = 2\pi\rho^2/\lambda_\mathrm{p}$, we obtain the following expression for the maximum pump power under the assumption that the pump and Stokes wavelengths differ little:

$$P_{\max} \approx \lambda_\mathrm{p}^2/(4n_2). \quad (2)$$

This expression for the maximum pump power essentially coincides (to within a dimensionless factor of 2) with the formula for the critical power for self-focusing. At this power, the calculated self-phase-modulation-induced spectral broadening of laser light (the ratio of the emission bandwidth at the output of the gain medium to that at its input) is ~2 [7]. This estimate correlates with experimental data reported by Losev et al. [8], which demonstrate that the spectral broadening of a pump pulse at the output of a gain medium is accompanied by a sharp drop in SRS efficiency.

In the case of transient SRS, the laser pulse energy should exceed the threshold energy E_th. Therefore, for SRS to occur in the case of the phase modulation of interacting waves, the following condition should be satisfied:

$$E_\mathrm{th}/\tau \leqslant P_{\max}, \quad (3)$$

where τ is the laser pulse duration.

Since the threshold laser pulse energy for SRS in a focused Gaussian beam is $E_\mathrm{th} = 45 T_2 \lambda_\mathrm{p}/g$ [9] (where g is the steady-state Raman gain coefficient), substituting this relation into (3) we find the minimum laser pulse duration:

$$\tau_{\min} \approx \frac{180 T_2 n_2}{g\lambda_\mathrm{p}}. \quad (4)$$

Note that, since $g \propto (\mathrm{d}\sigma/\mathrm{d}\Omega) N T_2 \lambda_\mathrm{p} \lambda_\mathrm{S}^2$ [10] (where $\mathrm{d}\sigma/\mathrm{d}\Omega$ is the differential spontaneous Raman scattering cross section and N is the gas concentration) and $n_2 \propto N n_2(1)$ (where $n_2(1)$ is n_2 at unit pressure), we have

$$\tau_{\min} \propto \frac{n_2(1)}{(\mathrm{d}\sigma/\mathrm{d}\Omega)\lambda_\mathrm{p}^2 \lambda_\mathrm{S}^2}. \quad (5)$$

Thus, the minimum pump pulse duration at which there is no backconversion is only determined by the ratio of the nonlinear refractive index to the spontaneous Raman scattering cross section and is independent of gas pressure.

Since the spontaneous Raman scattering cross section is related to the laser wavelength by $\mathrm{d}\sigma/\mathrm{d}\Omega \propto \lambda_\mathrm{p}^{-1}\lambda_\mathrm{S}^{-3}$ [10], the minimum possible pulse duration varies only slightly with laser wavelength:

$$\tau_{\min} \propto \frac{\lambda_\mathrm{S}}{\lambda_\mathrm{p}}. \quad (6)$$

Using relation (4) for evaluating the minimum pulse duration at a laser wavelength of 0.8 μm and nitrogen gas pressure of 10 atm, with $T_2 = 10$ ps, $n_2 = 3 \times 10^{-18}$ cm^2 W^{-1} and $g = 10^{-5}$ cm MW^{-1} [11], we obtain $\tau_{\min}^{\mathrm{N_2}} \approx 7$ ps. A similar estimate for hydrogen yields a shorter minimum laser pulse duration: $\tau_{\min}^{\mathrm{H_2}} \approx 1$ ps. These estimates were used to adjust the chirped laser pulse duration in our subsequent experiments.

3. Experimental study of SRS in compressed nitrogen

We performed experiments aimed at converting femtosecond Ti:sapphire laser pulses to the first Stokes wavelength via SRS in compressed nitrogen. Figure 1 shows a schematic of the experimental setup. We used an Avesta Project REUS-40F1K femtosecond Ti : sapphire laser system based on chirped-pulse amplification, with the following parameters: centre emission wavelength, 800 nm; bandwidth, 31 nm (see Fig. 3a); pulse duration, 35 fs; pulse energy, 2 mJ; 1/e^2 beam diameter, 9 mm; pulse repetition rate, 1 kHz. Varying the compressor grating separation, we ensured the generation of frequency-chirped pulses of increased duration at the output of the laser system.

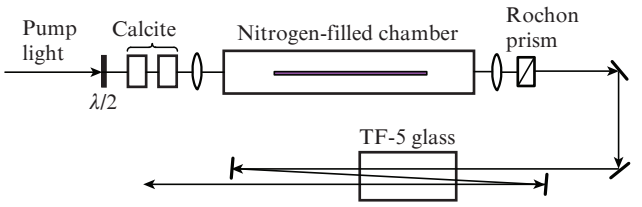

Figure 1. Configuration of the femtosecond compressed-nitrogen Raman laser.

To generate two sequential orthogonally polarised chirped laser pulses and vary the ratio of their energies, we used a system comprising two 1-cm-thick calcite plates and a half-wave plate [12]. These optical components allowed us to obtain two orthogonally polarised laser pulses separated by 10 ps.

The laser beam was focused by a lens with a focal length of 1.2 m onto the input end of a silica capillary tube 250 μm in inner diameter and 45 cm in length. The capillary was placed in a stainless steel chamber 1 cm in inner diameter and 1 m in length. The quartz windows of the chamber had antireflection coatings. The output beam was collimated by a lens and sent to a Rochon polarising prism. The prism was used to separate Stokes light from the delayed pump pulse.

In our experiments, we used negatively chirped laser pulses of ~10 ps duration. The Stokes pulse, which was also

negatively chirped, was compressed in time using a 26-cm-long TF-5 glass block. We used a configuration in which light made three passes through the glass block. The compressed pulse duration was measured by an Avesta Project ASF-20 autocorrelator.

In our first experiments, the chamber contained no capillary and the average laser output power was 2 W (pulse repetition rate of 1 kHz). Stokes light was generated at nitrogen pressures in the range 30–50 atm. Conversion efficiency, however, did not exceed 1%, and the spatial quality of the Stokes beam was not very high. We attribute these features of the conversion process to the development of optical nonuniformity around the focal waist as a result of the heating of the gas. This difficulty was eliminated by using a nitrogen-filled capillary, in which no gas convection occurred. The transmission of the capillary in the chamber under vacuum was measured to be 75%.

To optimise the working gas pressure, we measured conversion efficiency as a function of nitrogen pressure for the first Stokes at a wavelength of 980 nm under single-pulse pumping with a chirped pulse of ~10 ps duration. The results are presented in Fig. 2. The highest conversion efficiency, 16%, was reached at a nitrogen pressure of 20 atm. At higher pressures, conversion efficiency was lower because of the second Stokes generation at a wavelength of 1280 nm. In our subsequent experiments, the nitrogen pressure was 20 atm. It is worth noting that, when the chirped pulse duration was reduced to 2 ps, at none of the nitrogen pressures used was SRS detected.

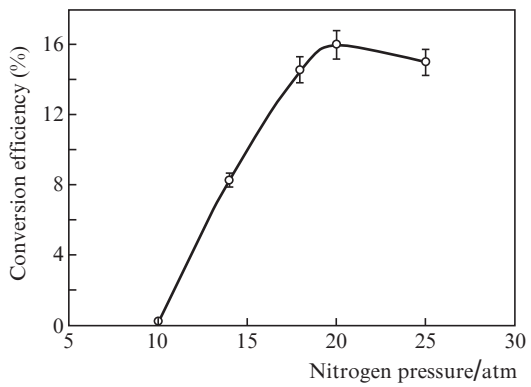

Figure 2. First Stokes (980 nm) generation efficiency as a function of nitrogen pressure.

As shown earlier [12], in the case of double-pulse pumping the most efficient and stable operation mode of a Raman converter is ensured when the energy of the first pump pulse is only slightly above the SRS threshold. This mode was ensured in the capillary-containing configuration (Fig. 1) at equal pump pulse energies. The ratio of the Stokes pulse energies was then 4:1, i.e. the energy of the delayed Stokes pulse was four times that of the Stokes pulse produced by the first pump pulse. Accordingly, at a total energy conversion efficiency of 16%, the efficiency of conversion into the broadband delayed Stokes pulse was ~12%. At pump pulse energies of 1 mJ, the energy of the delayed Stokes pulse was 0.25 mJ. The Stokes intensity profile across the output beam was nearly Gaussian.

Figure 3 shows Stokes pulse spectra obtained under single- and double-pulse pumping of the gain medium. It is seen that double-pulse pumping allows one to obtain Stokes pulses with a bandwidth exceeding that under single-pulse pumping by more than a factor of 2 (46 and 20 nm under double- and single-pulse pumping, respectively). Under single-pulse pumping with a negatively chirped pulse, the shortest wavelength spectral components, located at the leading edge of the pulse, are converted with low efficiency. As a result, the Stokes pulse bandwidth decreases.

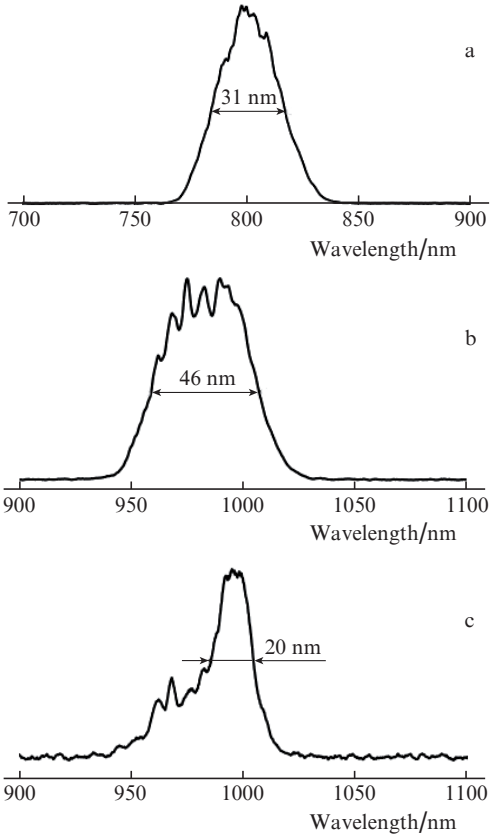

Figure 3. Output emission spectrum of the laser system (a) and Stokes pulse spectra under double-pulse (b) and single-pulse (c) pumping.

Pump pulse chirp reversal had no effect on the Stokes pulse bandwidth. This allows various Stokes pulse compressors to be used. In the case of positive chirp, grating or prism compressors can be used. Negatively chirped pulses can be compressed in a normal-dispersion transparent dielectric in its Stokes wavelength range. In this study, negatively chirped pulses were compressed in a TF-5 glass block. Figure 4 shows an autocorrelation function of a compressed pulse. Assuming a Gaussian pulse shape, we find that the pulse duration after the compression was 51 fs. The insufficient degree of compression (the duration of a transform-limited pulse with the spectrum shown in Fig. 3b is 38 fs) and the presence of subpulses are attributable to the inexact matching of the third and higher order dispersions in the case of pulse compression in a glass block [13].

4. Conclusions

The main results of the present study are as follows:

1. We have estimated the minimum possible pump laser pulse duration for the SRS process. Self- and cross-phase

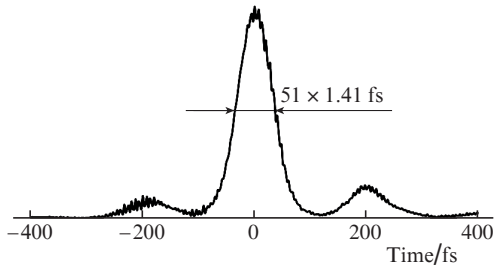

Figure 4. Autocorrelation function of a compressed Stokes pulse.

modulations of laser and Stokes waves have been shown to be determining processes. We have derived an analytical expression for the minimum possible pump pulse duration.

2. Femtosecond Ti : sapphire laser pulses have been converted to the first Stokes via vibrational SRS in nitrogen gas. Using a gas-filled capillary tube and exciting the active medium by two orthogonally polarised chirped laser pulses, we have reached an energy conversion efficiency of 12% and obtained 980-nm Stokes pulses of 51 fs duration at a repetition rate of 1 kHz and average pulse power of 0.25 W.

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