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NONLINEAR OPTICAL PHENOMENA

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Decreasing the amplitude of macroscopic quantum fluctuations in the case of transient SRS

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Abstract. **It is shown experimentally that, under the conditions of transient stimulated Raman scattering, pumping by two successive orthogonally polarised laser pulses makes it possible to reduce the amplitude of macroscopic quantum energy fluctuations of a Stokes pulse by a factor of 4 in comparison with single-shot pumping. An energy dispersion of 0.9% for the first Stokes component is obtained in hydrogen at a relative energy dispersion of ytterbium laser pulses of 0.4%.**

Keywords: *stimulated Raman scattering, femtosecond pulses, energy stability, quantum fluctuations.*

1. Introduction

The operational stability of a nonlinear laser beam converter is one of the main factors determining its application range. The most stringent requirements are generally imposed on the energy stability of a nonlinear converter. In particular, in the case of frequency conversion based on stimulated Raman scattering (SRS), of key importance is the Stokes energy stability (energy stability of Stokes pulse for pulsed SRS converters).

The SRS conversion of laser pulses shorter than few nanoseconds is most often performed under conditions of laser pulse single pass through an active medium. Schemes with single or multiple laser beam focusing in an active medium or various fibre systems (capillaries or fibres with multilayer photonic crystal cladding). As was shown in [1], one of the reasons for Stokes pulse energy instability in this case is the spontaneous nature of the Stokes photons initiating the SRS development. Quantum fluctuations of the number of initial Stokes photons lead to significant macroscopic fluctuations of Stokes pulse energy at the output of the active medium. It was established, both experimentally [2, 3] and theoretically [4], that macroscopic quantum fluctuations of Stokes pulse energy are maximum at a low conversion efficiency, when the Stokes pulse energy is much lower than the pump pulse energy. The amplitude of macroscopic quantum fluctuations decreases with an increase in conversion efficiency. Nevertheless, they are retained in the regime of saturated SRS conversion, when the laser and Stokes pulses have comparable energies [5, 6]. For example, according to the data of [6], the relative dispersion of Stokes pulse energy (ratio of the dispersion to the average pulse energy in percent) at a conversion efficiency of 40% amounted to 8%, which is close to the calculated values. Thus, macroscopic quantum fluctuations of Stokes radiation energy are an integral part of SRS conversion of laser pulses in schemes with Stokes radiation evolution from the level of spontaneous Raman scattering. In this context, the development of techniques aimed at reducing the influence of macroscopic quantum fluctuations and improving the operational stability of Raman converters is of high urgency.

In this work, we purpose and experimentally investigate a method for reducing the amplitude of macroscopic quantum fluctuations of Stokes pulse energy under conditions of transient SRS. A schematic diagram of the method is presented in Fig. 1. In the case of transient SRS, when the laser pulse width is shorter than the dephasing time T_2 of coherent oscillations in the active medium, a Stokes pulse with a power comparable with the pump pulse power arises after the laser pulse part carrying the threshold pump energy passes through the active medium [7]. In the case of laser pulse of fixed shape and energy, the instant of Stokes pulse occurrence will vary

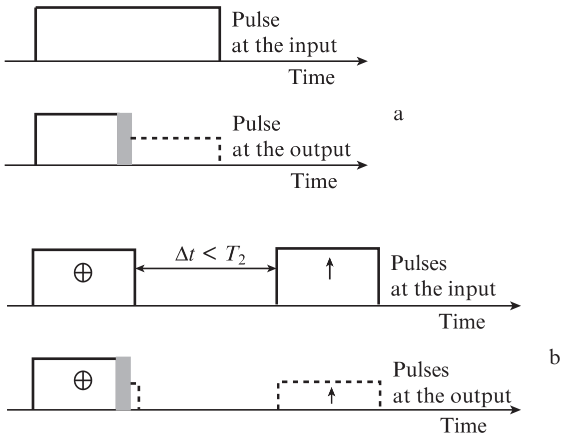

Figure 1. Schematic of the method for reducing macroscopic quantum fluctuations of Stokes pulse energy in the case of transient SRS: (a) single-shot pumping and (b) pumping by two orthogonally polarised pulses. The laser and Stokes pulses are shown by solid and dashed lines, respectively. The range of Stokes pulse instability is coloured grey.

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because of the quantum fluctuations of the initial number of Stokes photons. In turn, this will make the Stokes pulse energy fluctuate (Fig. 1a). To reduce the amplitude of macroscopic quantum fluctuations of Stokes pulse energy, it is proposed to divide the initial laser pulse into two subpulses spaced by a time interval shorter than the dephasing time T_2. Then, changing the ratio of subpulse energies or active-medium parameters, one must choose a conversion regime in which an intense Stokes pulse arises in the end of the first subpulse (Fig. 1b). Then the SRS conversion of the second subpulse will be performed with scattering from a dynamic polarisability grating (i.e., from coherent oscillations of the medium), formed by the first pulse, because the time interval between the pulses is shorter than the dephasing time T_2. Therefore, the spontaneous Raman scattering should not affect significantly the second subpulse conversion, and the quantum fluctuation amplitude should be much lower.

In the media with Raman scattering of scalar type, to which SRS from vibrational levels in gases belongs, the Stokes wave polarisation reproduces that of the pump wave. Therefore, to separate Stokes subpulses at the converter output, one can use a polarisation technique, in which orthogonal linearly polarised laser subpulses are formed at the input of the active medium.

2. Experimental results and discussion

The experiments were carried out using the optical scheme described in detail in [8, 9]. The laser radiation source was a femtosecond ytterbium laser (TETA, Avesta). The pulse repetition rate was 1 kHz. The pulse duration could be varied within 0.25–2 ps by frequency chirping. The centre radiation wavelength and spectral width were, respectively, 1033 and 8 nm. The maximum pulse energy reached 400 μJ. The relative dispersion of laser pulse energy amounted to 0.4%. To divide a laser pulse into two orthogonally polarised subpulses, the laser beam was transmitted through a birefringent calcite crystal placed before the converter chamber, which provided two orthogonally polarised pulses spaced by a time interval of 5 ps. The ratio of subpulse energies was varied by rotating a half-wave phase plate installed before the calcite crystal. The SRS conversion was performed in a 80-cm-long fused silica capillary with an inner diameter of 250 μm, placed in hydrogen-filled chamber 1 m long. At the chamber output, after selecting the Stokes radiation with a centre wavelength of 1.8 μm by a broadband filter, orthogonally polarised subpulses were separated by a Glan–Fresnel prism. An Ophir power meter with a pyroelectric sensor was used to measure the Stokes pulse energy and perform statistical data processing.

Before carrying out experiments with a double laser pulse, we measured the operational stability of single-shot-pumped Raman converter using a 1.6-ps single laser pulse. The hydrogen pressure was 28 atm. Figure 2 shows the dependences of the relative dispersion of Stokes pulse energy and the conversion efficiency on the laser pulse energy. The relative dispersion was measured for a sequence of 10^4 pulses. Several series of measurements were performed, based on which the average value of relative dispersion and measurement error were calculated. It can be seen in Fig. 2 that, in correspondence with the theory [4], the decrease in the pump pulse energy to the threshold level is accompanied by a sharp increase in the relative dispersion and deterioration of the operational stability of the Raman converter. A minimum relative dispersion (~4%) was obtained at a pump energy of about 350 μJ, at which the conversion efficiency approached its maximum value. A further increase in the pump energy did not lead to any significant increase in the conversion efficiency, whereas the relative dispersion increased to 5%. There may be two reasons for this increase: generation of the second Stokes component and transfer of Stokes radiation back to the pump wave [10]. Here, we did not analyse in detail the reasons for the increase in the relative dispersion at conversion efficiency saturation. However, in our opinion, the most likely process is the energy transfer from the Stokes wave back to the pump beam; this process occurs under conditions of efficient transient SRS conversion. In the case of efficient conversion, the Stokes radiation near the Stokes pulse trailing edge can be transferred to the radiation with a wavelength coinciding with that of the initial laser beam under conditions of anti-Stokes scattering from the polarisability grating recorded in the medium by the radiation of the pump beam and Stokes pulse leading edge. The phase-matching condition is automatically satisfied in this case, which leads to oscillations of Stokes pulse power and, as a result, reduces the operational stability. As for the generation of second Stokes component with a wavelength of ~8 μm, its generation threshold was not exceeded in our situation due to the smallness of the Raman gain (which is inversely proportional to the Stokes wavelength) and absorption of the second Stokes component in the fused silica capillary walls.

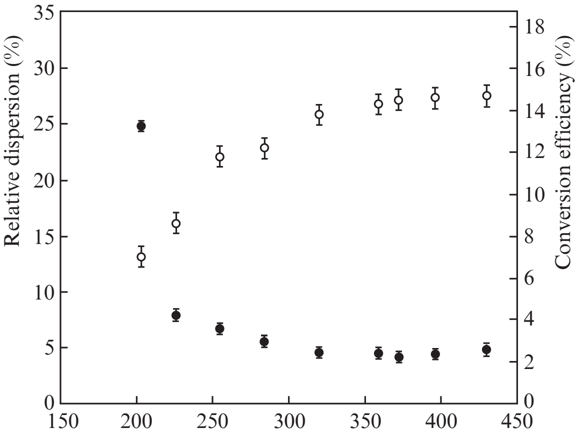

Figure 2. Dependences of (filled circles) the relative dispersion of Stokes pulse energy and (open circles) conversion efficiency on the single laser pulse energy.

The experiments with double-pulse excitation of the active medium were performed with subpulses having the same energy. As our previous experiments showed [9], this ratio of subpulse energies corresponds to the maximum values of the Raman gain increment and conversion efficiency. The subpulses were 1.6 ps wide, and the time interval between them was 5 ps. The double pulse energy was fixed at a level of 300 μJ. The hydrogen pressure in the converter chamber was varied to change the Raman gain increment (in the transient case the latter is proportional to the product of the laser pulse energy and particle concentration [7]).

Figure 3 shows dependences of the relative dispersion and fraction of Stokes pulse energy in the delayed subpulse on the hydrogen pressure in the chamber. These dependences indicate that, at low pressures (below 25 atm), when the Stokes component arises only in the delayed pump pulse, the relative dispersion exceeds 10%. An increase in hydrogen pressure leads to the following: the SRS energy threshold is exceeded even for the first pump subpulse, and scattering for the second pump subpulse begins on the previously formed wave of coherent oscillations of the active medium, because at a hydrogen pressure of 40 atm (maximum value reached in the experiment) the dephasing time T_2 is 120 ps [11]. The influence of quantum fluctuations of the number of seed Stokes photons decreases in this case, and the relative dispersion of the Stokes pulse energy is reduced. The most significant decrease in the relative dispersion (to 0.9% ± 0.4%, a value smaller by a factor of 4 than the minimum dispersion under single-shot pumping) was observed at a hydrogen pressure of 33 atm. In this regime, the first subpulse contains ~10% of the total Stokes pulse energy. Correspondingly, the second subpulse with a small relative dispersion contains 90% of the Stokes pulse energy. The total conversion efficiency to Stokes radiation was the same as for a single pump pulse: ~15%. An increase in hydrogen pressure to 40 atm reduces both the fraction and absolute value of Stokes energy in the delayed subpulse. The relative dispersion for the delayed pulse increases to 2.5%. As was noted above, we explain this effect by the unstable change in the Stokes pulse shape due to the Stokes radiation transfer back to the pump wave in case of saturated conversion under transient SRS conditions [10].

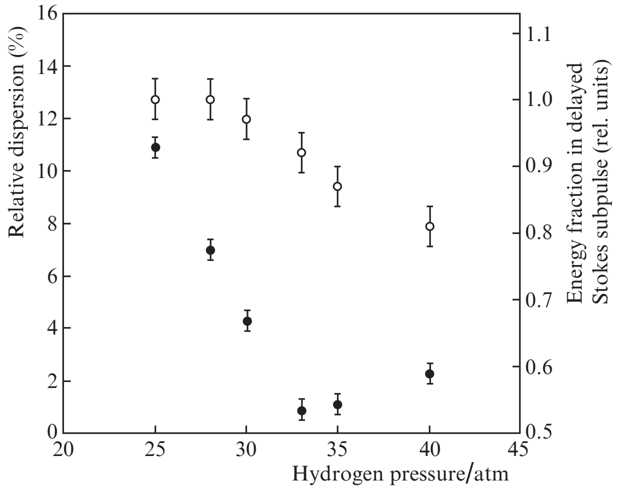

Figure 3. Dependences of (filled circles) the relative dispersion of delayed Stokes subpulse energy under double-pulse pumping and (open circles) the fraction of delayed Stokes subpulse energy in the total Stokes pulse energy on the hydrogen pressure.

It was shown in [8, 9] that Raman converters with double-pulse femtosecond pumping make it possible to implement (simultaneously with frequency shift) spectral broadening of the Stokes pulse due to the nonlinear cross- and self-phase modulation. The frequency-chirped Stokes pulse can then be compressed to a duration more than an order of magnitude shorter than that of the initial pump pulse. The operational stability of Raman compressor converter was not investigated. In this study, we considered the operational stability of a double-pulse-pumped Raman converter with a change in the subpulse duration, which varied from 400 fs to 1.8 ps. At shorter durations the Raman conversion efficiency sharply decreased due to the effects of nonlinear phase modulation [12]. The hydrogen pressure was 35 atm, and the double pump pulse energy was 250 μJ. The measurement results are presented in Fig. 4. Note that, in the case of transient SRS, the relative dispersion of the total Stokes pulse energy coincides with the dispersion under pumping by a single pulse having an energy equal to the total double pulse energy. The relative dispersion of Stokes pulse energy increases with a decrease in the subpulse duration. This dependence may be related to the decrease in the Raman gain increment due to the nonlinear phase modulation [12]. A decrease in the Raman gain increment, as well as a decrease in the laser pulse energy, leads to an increase in the relative dispersion (Fig. 2). Hence, for the shortest subpicosecond pulses, we could reduce the relative dispersion of Stokes pulse energy only by half as compared with single-shot pumping.

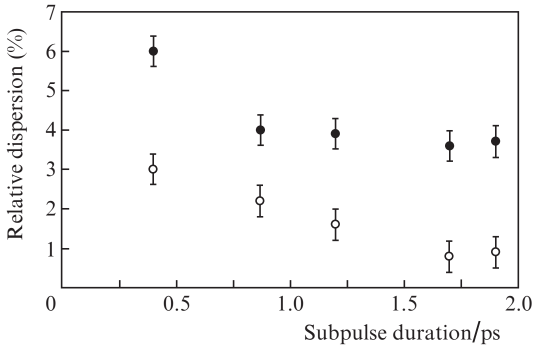

Figure 4. Dependences of (filled circles) the relative dispersion of the total energy of two Stokes subpulses and (open circles) the relative dispersion of the energy of delayed Stokes subpulse on the subpulse duration.

To conclude, we should note that the proposed scheme of a double-pulse-pumped Raman converter [9] is promising for designing frequency converters with operational stability close to that of the laser system. The application of this scheme with fibres of larger diameter and higher power ytterbium lasers [13] opens ways to generate highly stable millijoule pulses in the vicinity of 2 μm for mid-IR nonlinear optics experiments.

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