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SRS generation of femtosecond pulses in a methane-filled revolver hollow-core optical fibre

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Abstract. **The influence of the width of pump pulses (with λ_p = 1.026 μm) on the SRS generation of femtosecond pulses in a methane-filled hollow-core fibre is investigated. The width of pump pulses is controlled by transmitting them through an optical dispersion-managed system and thus making them linear frequency-modulated (chirped). Regimes of both the preferred SRS generation of femtosecond pulses at the Stokes wavelength (λ_{st} = 1.464 μm) and the generation of a multiband supercontinuum are experimentally demonstrated. A quantum efficiency of 41 % and a pulse width of 590 fs at the Stokes wavelength are obtained for the 1.026 → 1.464 nm Raman generation.**

Keywords: *fibre laser, hollow-core fibre, revolver fibre, ultrashort pulse, stimulated Raman scattering.*

1. Introduction

Femtosecond lasers are used to solve many scientific and applied problems [1,2]. Hence, it is of great importance to expand the spectral range of available femtosecond laser sources. In particular, an urgent task is to pass from the well-mastered wavelength range $\lambda \approx 1$ μm (ytterbium femtosecond lasers) to the longer wavelength spectral region for future mastering the mid-IR range.

One of the long-wavelength conversion methods is stimulated Raman scattering (SRS). However, this technique meets some difficulties in the case of femtosecond pump pulses. Since SRS has a certain inertia, its efficiency is suppressed by almost instantaneous nonlinear effects (due to the electronic polarisability) when passing to femtosecond pulses. In particular, it is known that the main nonlinear effects competing with SRS are self-phase modulation (SPM) and self-focusing [3].

A method for suppressing nonlinear processes competing with SRS under pumping by femtosecond pulses was demonstrated for the first time in [4]. This method is based on linear frequency modulation (chirp), which transforms the pump pulse duration from femtosecond to picosecond range. Then efficient SRS is implemented, in which the linear chirp of pump pulses is transferred to the Stokes pulses. In the final stage the pulse at the Stokes wavelength can be compressed again to femtosecond duration. The above-described method was developed in several studies, where both solid-state [5] and gaseous Raman-active media [6–9] were analysed. SRS amplification of chirped picosecond pulses with their subsequent compression to subpicosecond duration was also demonstrated in [10].

SRS on vibrational transitions in light molecular gases is of particular interest, because it provides large Stokes shifts (4155, 2991, and 2917 cm^{-1} for H_2, D_2 and CH_4 molecules, respectively). The number of studies devoted to the light conversion in gaseous media has increased multiply after designing hollow-core fibres (HCFs). Due to the small mode field diameter (~5–50 μm) and large interaction length (~1–10 m), hollow-core fibres made it possible to reduce the thresholds of nonlinear effects by several orders of magnitude in comparison with the case of light focusing in the bulk of a gaseous medium. In addition, an important distinction of fibres from bulk media is the presence of waveguide dispersion, which may affect significantly the regime of femtosecond pulse conversion in a gas-filled fibre.

To date, various regimes of femtosecond pulse propagation in hollow-core fibres have been demonstrated. In particular, many studies were devoted to the generation of dispersion waves in the UV range [11–13], the time-dependent SRS on transitions in molecular gases [14], and the supercontinuum generation [15]. Recently researchers have actively investigated the influence of pump pulse duration on the SRS in gas-filled hollow-core fibres in order to implement femtosecond-pulse generation at the Stokes wavelength [16,17]. Nevertheless, the regimes of SRS generation of femtosecond pulses with suppression of competing nonlinear processes in gas-filled fibres remain scarcely studied.

In this paper, we report the results of studying the influence of the pump pulse duration (λ_p = 1.026 μm), controlled by linear frequency modulation, on the SRS generation of femtosecond pulses in a methane-filled hollow-core fibre. It is shown that the SRS is the dominant nonlinear effect (allowing one to convert deliberately the pump radiation into the radiation at the Stokes wavelength) for transform-limited pulses with a duration of about 200 fs, time-extended to a duration τ of more than 1.5 ps. The 1.026 → 1.464 nm conversion with a quantum efficiency of 41 % was demonstrated for $\tau \geqslant 1.5$ ps. The minimal pulse duration at the Stokes wavelength was found to be 590 fs. At the same time, at $\tau < 1.5$ ps, the dominant effect is the SPM, which leads (jointly with the SRS, four-wave parametric generation, and cross-phase modulation) to the generation of a multiband supercontinuum.

2. Experimental

A schematic of the experimental setup used to investigate the influence of chirped pump pulse duration on the SRS genera-

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tion of femtosecond pulses in a gas-filled hollow-core fibre is shown in Fig. 1. The pump source was a PHAROS femtosecond laser (Light Conversion), which generated transform-limited pulses with a duration $\tau_0 \approx 210$ fs and energy up to 200 μJ at a wavelength of $\lambda_p = 1.026$ μm. The pulse duration τ could be varied in the range of 0.2–10 ps by a built-in compressor, which provided both positive and negative frequency modulation with a factor $|C| = \sqrt{(\tau/\tau_0)^2 - 1} \leqslant 50$.

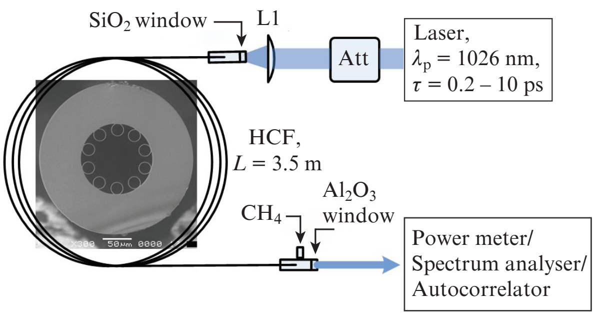

Figure 1. Schematic of the experimental setup: (Att) attenuator; (L1) focusing lens; (HCF) hollow-core fibre. Input (SiO_2) and output (Al_2O_3) windows and a gas (CH_4) feed unit are indicated. The inset shows a SEM image of a fibre end face.

The radiation power was controlled by an external attenuator. The radiation transmitted through it was fed into a hollow-core fibre using a lens L1 ($f = 200$ mm). The input efficiency was 94%. The fibre in use (Fig. 1, inset), which had a length of 3.5 m (in a coil 50 cm in diameter), was a revolver-type HCF [18], whose core was filled with methane under a pressure of 15 atm. The core diameter was 80.7 μm, a value corresponding to the fundamental mode field diameter 58.2 μm at the pump wavelength. The fibre cladding was formed by ten silica glass capillaries; their wall thickness and external diameter were 1.3 and 22.2 μm, respectively. The fibre ends were hermetically glued into miniature vacuum chambers (cells). The input and output cells had silica glass (transmittance $T_{SiO_2} = 0.92$) and sapphire ($T_{Al_2O_3} = 0.844$) windows for radiation input/output, respectively. The radiation from the fibre output end face was fed into an optical spectrum analyser, an autocorrelator, or a power meter. When measuring the Stokes component ($\lambda_{st} = 1.464$ μm), the pump ($\lambda_p = 1.026$ μm) power was filtered off using a silicon plate.

The optical loss spectrum for the fibre fundamental mode was calculated using the COMSOL Multiphysics software. The fundamental mode losses at wavelengths of 1.026 and 1.464 μm were theoretically estimated as 1.2 and 12 dB km^{-1}, respectively (Fig. 2). The material losses caused by the absorption on vibrational–rotational transitions in methane molecules at a temperature $T = 296$ K and a pressure $p = 15$ atm were found to be 0.15 dB m^{-1} ($\lambda_p = 1.026$ μm) and 0.53 dB m^{-1} ($\lambda_{st} = 1.464$ μm) [19].

3. Results and discussion

To reveal the nonlinear effects that are dominant when chirped pulses propagate in a methane-filled fibre, we investigated the spectral composition of light at the fibre output.

In the case of transform-limited input pulses ($\tau_0 = 207$ fs, $\lambda_p = 1.026$ μm), an increase in the input pulse energy to $E_p \geqslant 1$ μJ (which corresponds to the intensity $I_p \geqslant 2 \times 10^{11}$ W cm^{-2})

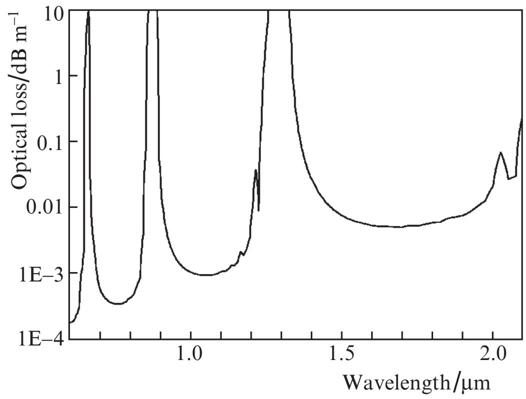

Figure 2. Optical loss spectrum of the revolver fibre under study, calculated by numerical simulation. The absorption caused by the presence of methane is omitted.

led primarily to broadening of the output spectrum at the pump wavelength (Figs 3a–3c). When the pulse energy E_p reached a level on the order of 1 μJ, we observed an SRS threshold, accompanied by the generation of Stokes radiation in the vicinity of about 1.464 μm (Fig. 3c). The wavelength $\lambda_{st} = 1.464$ μm corresponds to the pump radiation ($\lambda_p = 1.026$ μm) scattering from symmetric vibrations of C–H bonds in methane molecules ($\Omega_R = 2917$ cm^{-1}). It can be seen that the Stokes spectrum is significantly broadened even at the SRS threshold. With a further increase in the input pulse

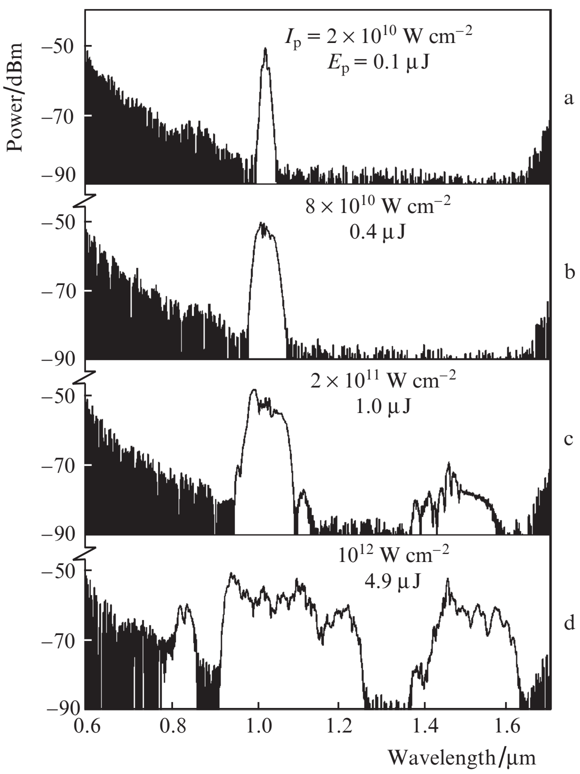

Figure 3. Spectra measured at the output of a revolver fibre filled with methane at room temperature and pressure $p = 15$ atm, under pumping by transform-limited pulses ($\tau_0 = 207$ fs, $\lambda_p = 1.026$ μm) with different energies and intensities.

energy E_p, one could observe a much more significant spectral broadening, which passed into multiband supercontinuum generation at $E_p \geqslant 5$ μJ ($I_p \geqslant 10^{12}$ W cm^{-2}) (Fig. 3d) [20]. Note that the dips in the output spectrum, observed near the wavelengths $\lambda \approx 0.9$ and 1.3 μm in the supercontinuum generation regime (Fig. 3d), correspond to the regions of high optical loss in the transmission spectrum of a hollow fibre (see Fig. 2). The long-wavelength edge of the supercontinuum ($\lambda \approx$ 1.62 μm) is explained by the absorption in methane, whose molecules have dipole-active transitions with absorption coefficients of 4–440 dB m^{-1} (at a pressure p = 15 atm and a temperature T = 296 K) in the spectral range $\lambda \approx$ 1.62–1.84 μm [19].

The influence of the duration of frequency-modulated (chirped) pump pulses on their propagation regime was studied at a pump energy E_p = 2 μJ introduced into a methane-filled fibre. The output radiation spectra revealed the presence of two propagation regimes (Fig. 4). The vibrational SRS is dominant at a chirped pulse duration $\tau \geqslant 1.5$ ps ($I_p \leqslant 5 \times 10^{10}$ W cm^{-2}), which is valid for both negative and positive values of the frequency-modulation factor C (Figs 4a and 4e). With a decrease in the chirped pump pulse duration ($\tau <$ 1.5 ps), the SRS is accompanied by a significant spectral broadening, which is observed near both the pump and Stokes wavelengths (Figs 4b and 4d). We primarily relate this spectral broadening to the influence of self-phase modulation, whose contribution is maximal for transform-limited input pump pulses (Fig. 4c).

Thus, the study of the spectral composition of output radiation showed that, increasing the pump pulse duration from τ_0 = 207 fs ($I_p \approx 4 \times 10^{11}$ W cm^{-2}) to values above 1.5 ps ($I_p \approx 0.5 \times 10^{11}$ W cm^{-2}), one can convert deliberately pump radiation into radiation at the Stokes wavelength due to the SRS on vibrations of methane molecules.

The dependences of the durations of input and output pulses on the pump-pulse chirp factor $|C| = \sqrt{(\tau/\tau_0)^2 - 1}$ are presented in Fig. 5. It can be seen that the durations of the pump pulses at the output and input of hollow-core fibre barely differ. This circumstance indicates that the waveguide dispersion hardly affects the propagation of pulses with a wavelength of 1.026 μm in fibres about 3.5 m long. At the same time, the duration of output Stokes pulses is smaller than the pump pulse duration by a factor of about 2.5. Apparently, the reason is that the energy of the pump-pulse leading edge is spent on the excitation of molecular vibrations and, correspondingly, is not converted into Stokes pulses.

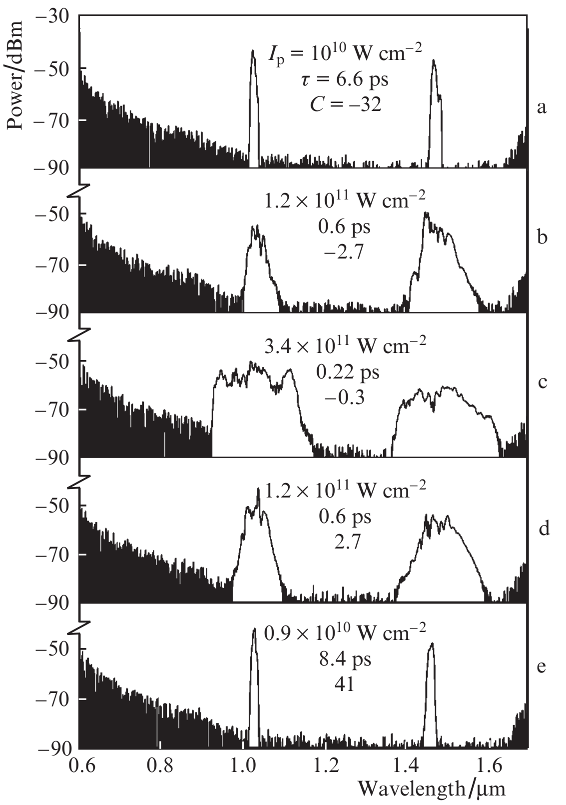

Figure 4. Spectra measured at the output of a revolver fibre filled with methane at room temperature and pressure p = 15 atm, under pumping by linearly chirped pulses (λ_p = 1.026 μm, E_p = 2 μJ) with different intensities.

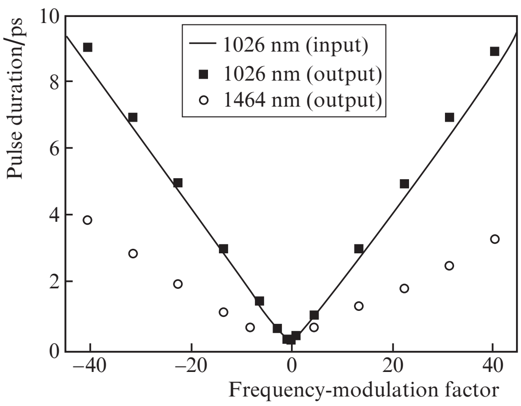

Figure 5. Dependences of the pulse duration on the frequency-modulation factor of pump pulses for the (solid line) input and (squares) output pump pulses and (circles) output pulses at the Stokes wavelength.

Note that, in the case of chirped pump pulses, a decrease in the pulse duration at SRS conversion is accompanied by the narrowing of the spectrum at the Stokes wavelength. This effect hinders subsequent Stokes pulse compression to a duration corresponding to the initial transform-limited pump pulse. To solve this problem for non-waveguide light propagation in a bulk medium, special procedures must be developed [7]. However, as can be seen in Figs 4b and 4d, joint SRS + SPM regime can be implemented in hollow-core fibres. Under these conditions, the spectral width of Stokes pulses exceeds the pump spectrum width, which can be used for subsequent compression of these pulses.

Note also that, at a pump pulse duration exceeding 1.5 ps (λ_p = 1.026 μm, $I_p < 5 \times 10^{10}$ W cm^{-2}), when the dominant nonlinear effect is SRS, the autocorrelation function (ACF) of Stokes pulses (λ_{st} = 1.464 μm) is described adequately by a Gaussian. The characteristic ACF for τ_1 = 1.7 ps ($I_p \approx 4.4 \times 10^{10}$ W cm^{-2}, C = –8) is shown in Fig. 6. The Stokes pulse duration was found to be 590 fs in this case.

The most efficient 1.026 → 1.464 μm conversion was observed when the duration of chirped pump pulses at λ_p = 1.026 μm corresponded to the regime of SRS dominance over SPM ($\tau \geqslant 1.5$ ps, $|C| \geqslant 7$). A characteristic dependence of the quantum efficiency on the pump pulse energy satisfying this condition is presented in Fig. 7 for a pump pulse duration τ = 3.6 ps (C = –17). The maximum quantum efficiency amounted to 41%; this value was obtained under pumping a fibre by

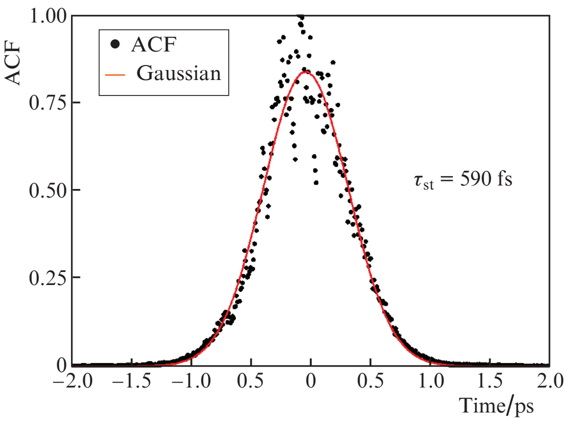

Figure 6. Autocorrelation function of output pulses at λ_{st} = 1.464 μm under pumping a hollow fibre by chirped pulses with a duration τ = 1.7 ps (λ_p = 1.026 μm, $I_p \approx 4.4 \times 10^{10}$ W cm^{-2}, $C = -8$).

6 μJ pulses ($I_p \approx 6 \times 10^{10}$ W cm^{-2}). A further increase in the pump pulse energy reduced the quantum efficiency; we relate this reduction to the absorption by methane molecules at the Stokes wavelength (absorption coefficient 0.53 dB m^{-1} at p = 15 atm). When the pump pulse duration was much shorter than 1.5 ps, the quantum SRS efficiency decreased essentially. This fact is explained by the influence of the self-phase modulation, which significantly broadens the spectrum. As a result, energy loss occurs in high-loss regions of the hollow fibre (see Fig. 2) and within the methane absorption bands (at λ > 1.62 μm).

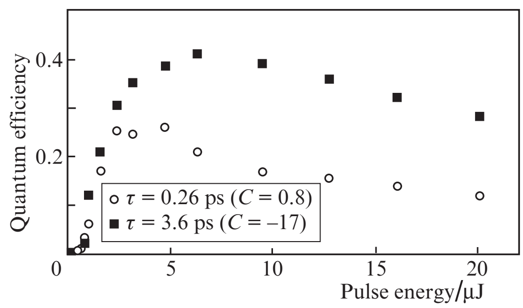

Figure 7. Dependences of the SRS quantum efficiency in a fibre filled with methane at a pressure of 15 atm on the energy in chirped pump pulses (λ_p = 1.026 μm) with durations of 0.26 and 3.6 ps.

4. Conclusions

It was shown experimentally that the duration of femto- and picosecond pump pulses, varied by means of linear frequency modulation, makes it possible to control the competition between nonlinear SRS and SPM effects in gas-filled hollow-core fibres. Two regimes of chirped pulse (λ_p = 1.026 μm) conversion were demonstrated by an example of a revolver fibre filled with methane at a pressure of 15 atm. These were SRS generation of femtosecond pulses at the Stokes wavelength (λ_{st} = 1.464 μm) and generation of a multiband supercontinuum. A quantum efficiency of 41% and a minimum Stokes pulse duration of 590 fs were obtained under conditions of 1.026 → 1.464 μm SRS generation. These results indicate good prospects of gas-filled revolver fibres for efficient SRS generation of femtosecond pulses.

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