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1.56-to-2.84 μm SRS conversion of chirped pulses of a high-power erbium fibre laser in a methane-filled hollow-core revolver fibre

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Abstract. **Single-cascade 1.56-to-2.84 μm SRS conversion is demonstrated in a hollow-core revolver fibre filled with methane at a pressure of 25 atm under pumping by positively chirped pulses of a high-power erbium-doped all-fibre laser. At a maximum pump pulse energy of 34 μJ (average power 3.74 W) and a pump pulse duration of about 260 ps, ultrashort pulses (USPs) with a duration of 110 ps and an energy of 1.33 μJ (average power 133 mW) are achieved at the centre wavelength of 2.84 μm. The gas fibre Raman lasers based on hollow-core fibres with pumping by high-power fibre sources are promising for producing all-fibre systems emitting USPs in the mid-IR range.**

Keywords: *chirped pulse, stimulated Raman scattering, gas fibre laser, mid-IR range, hollow-core fibre, erbium-doped fibre amplifier.*

Sources of coherent radiation in the spectral range 2.5–5 μm attract much attention because they are promising for application in telecommunication and defence fields, as well as in medicine and for precision gas analysis [1–3]. In particular, the spectral range 2.7–3 μm, which includes the intense absorption line of the OH bonds, is of interest for biomedical applications.

Lasers emitting ultrashort pulses (USPs) of pico- and femtosecond durations in the mid-IR range are of particular interest. Owing to the characteristic broad spectrum consisting of narrow equidistant lines (optical comb), these sources are required in ultrahigh resolution spectroscopy [4], for generating terahertz radiation [5], and in minimally invasive microsurgery [6]. In addition, the subpicosecond duration of laser pulses makes them indispensable for investigating fast intramolecular processes and for microprocessing of various materials (including transparent ones).

At present, there exist several main approaches to the development of USP laser sources in the spectral range 2.5–3 μm based on the use of solid-state, fibre, and hybrid architectures of the corresponding laser systems. The first ones include sources based on vibronic single and polycrystals of chromium-doped transition metal chalcogenides [7], which emit high-power subpicosecond pulses at wavelengths around 2.5 μm, as well as parametric oscillators and amplifiers [8], which allow wavelength conversion in wider ranges. However, along with the necessity to fulfil the phase-matching conditions in the used nonlinear crystals, parametric oscillators have a rather low efficiency and insufficiently high output beam quality.

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Recently, USP fibre lasers and amplifiers have been actively developed based on fluoride glass fibres (ZrF_4, InF_3, and ZBLAN) doped with various rare-earth ions (Er^{3+}, Dy^{3+}, Pr^{3+}, and Ho^{3+}) [9]. Unfortunately, soft fluoride glass demonstrates very low radiation resistance and mechanical strength in comparison with silica glass, which restricts the USP energy and peak power [9] and, as a result, hinders the use of fluoride fibres in high-power laser systems competitive with solid state analogues.

Another promising method for generation of pico- and femtosecond pulses in the mid-IR range is stimulated Raman scattering (SRS) in gaseous media due to a large vibrational Stokes shift in light gases [for example, the Stokes shift is 2917 cm^{-1} for methane (CH_4), 2987 cm^{-1} for deuterium (D_2), and 4155 cm^{-1} for hydrogen (H_2)] in the case of pumping by widely used USP sources in the near-IR range [10, 11]. This method served as a basis of the concept of Raman gas lasers and was initially implemented using cell and capillaries filled with Raman-active gases [10, 11]. To date, SRS generation of USPs with a minimum duration of 20 fs and an energy of 30 μJ at the centre wavelength of 1.8 μm was achieved with an efficiency of ~12% in a hydrogen-filled capillary under pumping by 400-fs pulses of a high-power ytterbium hybrid source of radiation with a wavelength of 1030 nm [12]. Note that generation of USPs at longer wavelengths via SRS in gas-filled capillaries has not yet been obtained.

Significant progress in the development of mid-IR Raman lasers is related to the appearance of silica revolver hollow-core fibres (HCFs) [13], which have low (~0.1 dB m^{-1}) losses in the near-IR (1–1.5 μm) spectral range and quite acceptable (~1 dB m^{-1}) losses in the range 2.5–5 μm [13], which are 1000–4000 times lower than the losses in silica glass itself. The use of HCFs makes it possible to considerably decrease the SRS threshold in the steady-state regime due to a decrease in the mode area and an increase in the interaction length [13]. As a result, Raman gas fibre lasers (GFLs) based on silica HCFs can be pumped by fibre lasers with nanosecond pulse durations [13–15], which, in turn, opens the way to the development of all-fibre pulsed sources of IR radiation [16].

The physical picture of SRS in gas-filled HCFs becomes much more complicated in the case of conversion of optical combs of pico- and femtosecond pulses due to, in particular, the influence of dispersion in HCFs [17]. Moreover, in the case of conversion of USPs with duration τ_p corresponding in the order of magnitude to the dephasing time in gas T_2 (a more exact condition is given by the inequality $\tau_p < 20T_2$ [10]), the SRS regime becomes nonstationary [10–13, 17–19], because of which its dynamics depends on the pump pulse energy more strongly than on the peak power, which is especially clearly seen in the case of $\tau_p \ll T_2$, when SRS becomes significantly nonstationary [10–12, 18, 19]. The SRS gain in the transient regime decreases, and the SRS threshold increases with decreasing duration of chirped pump pulses and saturates under the conditions of strong nonstationarity [19]. In addition, we should take into account that, other conditions being equal, the Raman gain decreases with increasing radiation wavelength [10]. These factors hinder the use of the USP fibre laser system for pumping mid-IR Raman GFLs.

By now, only two papers [20, 21] have reported on successful operation of Raman GFLs in the spectral range 2.5–3 μm under pumping by high-power USPs from, in both cases, solid-state ytterbium lasers. In [20], the second-order Stokes pulses with a duration of less than 12 ps and a maximum energy of 113 μJ were obtained at a wavelength of 2.812 μm in a methane-filled revolver HCF 3 m long pumped by 12-ps transform-limited pulses at a wavelength of 1064 nm. In [21], the duration of second-order Stokes pulses at a wavelength of 2.68 μm was 920 fs (which is the first demonstration of subpicosecond mid-IR Raman GFLs) in the case of pumping of a deuterium-filled revolver HCF 2.9 m long by 10-ps chirped USPs. It should also be noted that the threshold pump pulse energy in both cases exceeded 20 μJ (~26 [20] and ~50 μJ [21], respectively).

The aim of the present work was to demonstrate the possibility of using an all-fibre USP source for pumping a Raman gas laser emitting in the spectral range 2.5–3 μm. To solve this problem, we chose the single-cascade 1.56-to-2.84 μm SRS conversion in a methane-filled revolver HCF under pumping by USPs of an erbium fibre laser, which, due to the combination of parameters, seems to be the most promising scheme.

An important advantage of methane is the absence of rotational components in the Raman spectrum, which considerably reduces the polarisation sensitivity of SRS conversion. In addition, the dephasing time in methane ($T_2 = 17$ ps at the pressure $p = 25$ atm [10]) is much shorter than in the other Raman-active light gases (hydrogen and deuterium) at comparable SRS gains [10], which leads to a noticeable decrease in the SRS threshold (to units of microjoules) in methane GFLs at the 1.46-μm Stokes component under pumping by picosecond chirped USPs at a wavelength of 1.026 μm [22]. These data make methane a preferable active medium of Raman GFLs pumped by fibre USP sources.

In turn, it is necessary to note that SRS conversion of high-power laser pulses in methane was reported for the first time in 1963 by Minck et al. in [23], in which a gas cell 15 cm long was pumped by ruby laser pulses with a duration of about 30 ns. A rather detailed review of achievements in the field of cw and pulsed SRS both in various Raman-active gases and in crystals and fibres is presented in [24–26]. We should also note that work [10] mentioned above provides a comprehensive comparison of various SRS parameters in capillary fibres and cells filled with methane, deuterium, and hydrogen under pumping by a picosecond dye laser with the central wavelength of around 560 nm. However, it is fibre USPs sources whose use for pumping HCF Raman gas lasers may open the way to the development of all-fibre mid-IR USP laser systems, which is a very topical problem.

The erbium-doped all-fibre USP source was fabricated using a well-proven method of amplification of chirped pulses. 250-fs Gaussian pulses of an erbium master oscillator (centre wavelength 1.56 μm) [27] were stretched to a duration of ~1 ns and attained a positive frequency modulation (chirp) in a fibre–stretcher with a positive group velocity dispersion and a length of about 500 m. Next, the pulses were amplified in an erbium-doped fibre pumped into the core by a single-mode diode, and then their repetition rate was decreased with the use of an acousto-optic shutter. The pulse repetition rate was decreased from 38.1 MHz to 100 kHz. The low-repetition-rate pulses were sent to a high-power hybrid amplifier based on erbium-doped fibres with an increased mode area and multimode diode pumping [28], which completed the formation of chirped USPs of the erbium-doped all-fibre pump source.

The collimated output radiation of the high-power erbium laser was coupled into a revolver fibre 2.8 m long with a hollow core 75 μm in diameter, which was surrounded by ten noncontacting silica capillaries with walls 1.15 μm thick [21]. The scanning electron microscope image of the fibre end face is shown in Fig. 1a. The optical losses in the fibre in a wide wavelength range calculated using the COMSOL software are presented in Fig. 1b.

The loss spectrum has a band structure typical for revolver HCFs [13]. The loss at the pump wavelength ($\lambda_p \approx 1.56$ μm), which almost completely coincides with the centre of the corresponding HCF transmission band, are negligibly small and

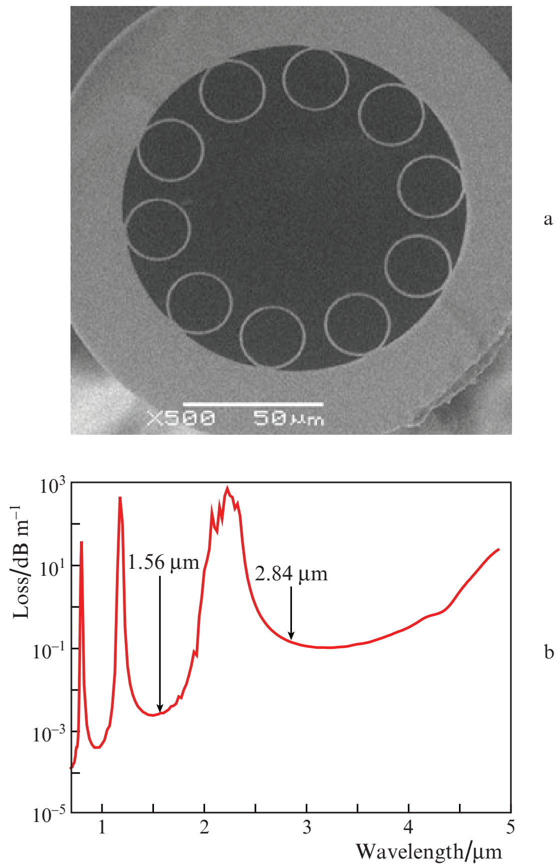

Figure 1. (Colour online) (a) Image of the HCF end face and (b) optical loss spectrum of this fibre.

do not exceed 2.5 dB km^{-1}, while the losses in the region of the Raman GFL wavelength near 2.84 μm comprise an acceptable value of 0.13 dB m^{-1}.

The ends of the hollow-core fibre were glued into special hermetic cells with transparent input (silica) and output (sapphire) windows. The Raman-active gas (in our case, methane) was injected through the output cell, while the input cell was mounted on a precision adjusting stage with five degrees of freedom to optimise the pump radiation coupling into the hollow-core fibre. The output radiation from the HCF was collimated by a zinc–selenium plano-convex lens and directed to the measuring part of the setup, which consisted of an MDR-24 monochromator with a semiconductor photodetector (working spectral range 1–5 μm), a scanning autocorrelator (maximum delay time 850 ps), and an average-power meter. To separate the Stokes pulse and suppress the pump pulse during the measurements, we used a broadband filter based on a germanium plate antireflection coated for the range 1.9–5 μm. The experiments were performed at a methane pressure of 25 atm.

In the course of experiments, we achieved Raman lasing in the mid-IR spectral range. The SRS threshold in the methane-filled HCF was observed at an average output power of the pump erbium fibre laser of 2 W, which corresponds to a chirped USP energy of 18 μJ. An increase in the average pump energy (and in the USP energy) up to 3.74 W (or 34 μJ) was accompanied by an increase in the SRS average power up to the maximum value equal to 133 mW, after which this increase ceased. The energy saturation is probably caused by losses in the gas-filled HCF at the Stokes wavelengths [13, 15, 22].

The spectrum of the SRS pulse at its maximum power and the corresponding pump pulse spectrum at the HCF output are presented in Fig. 2a. It should be noted that the central Stokes wavelength ($\lambda_S \approx 2.84$ μm) corresponds to the Raman shift in methane (2917 cm^{-1}) for the pump wavelength $\lambda_p \approx$ 1.56 μm. At the same time, apart from the pump and the first Stokes component, the wavelength range 1–3 μm contains no other spectral lines, which is typical for SRS in methane.

Figure 2b shows the autocorrelation functions (ACFs) of the Stokes pulse intensity at the HCF output and of the output chirped pulse of the erbium fibre pump source. From the Gaussian approximation of the measured ACFs, the 110-ps Stokes pulse was found to be 2.4 times shorter than the corresponding pump pulse (average duration 260 ps), which is typical for SRS conversion of chirped pulses. Indeed, in the transient SRS regime (which is observed in our case because the $\tau_p < 20T_2$ and $\tau_p > T_2$ conditions are fulfilled simultaneously), only that fraction of the pump pulse (and the corresponding spectral components) is converted for which the SRS energy threshold is exceeded, which inevitably leads to the Stokes pulse narrowing [11, 12, 18–22]. This means that, in the case of a positively chirped pump pulse, the short-wavelength spectral components at the pump pulse trailing edge are converted. The jagged shape of the pump pulse ACF is caused mainly by nonlinear effects in the output amplification cascade of the high-power erbium fibre laser at high USP energies.

Taking into account the efficiency of pump coupling into the HCF (~70%), the maximum SRS quantum efficiency with respect to the coupled pump radiation is ~10%, ant the maximum Stokes pulse energy in this case reaches 1.33 μJ. Since the linear pump pulse chirp upon SRS is directly transferred to the Stokes pulse [11, 12, 18, 21], the latter can be compressed by, for example, an external grating compressor

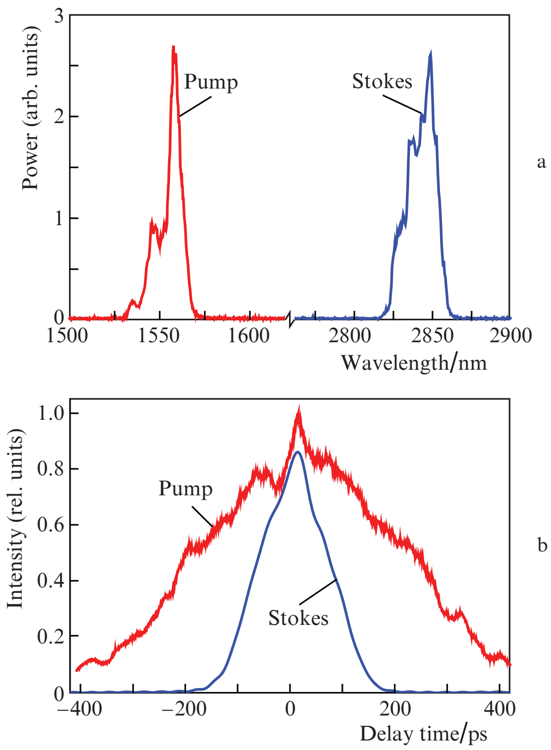

Figure 2. (Colour online) (a) Spectra and (b) autocorrelation functions of the pump and Stokes pulses.

to a duration of ~1 ps; based of the width of its spectrum (at a level of –3dB), we have $\Delta\lambda_S \approx 20$ nm ($\Delta\nu \approx 25$ cm^{-1}).

Thus, we have demonstrated for the first time the single-cascade 1.56-to-2.84 μm SRS conversion in a revolver hollow-core fibre filled with methane at a pressure of 25 atm under pumping by positively chirped pulses of a high-power erbium all-fibre laser source. In the future, we intend to study in more detail the parameters of the developed Raman GFL with varying methane pressures and hollow-core fibre lengths, as well as to use other Raman-active gases (hydrogen and deuterium). It should also be emphasised that the concept of Raman gas fibre lasers based on hollow-core fibres and pumped by high-power fibre USP sources is promising from the point of view of development of all-fibre mid-IR USP laser systems.

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References

1. Jackson S.D. *Nat. Photonics*, **6** (7), 423 (2012).
2. Bekman H.H.P.Th., van den Heuvel J.C., van Putten F.J.M., Schleijpen R. *Proc. SPIE*, **5615**, 27 (2004).
3. Werle P., Slemr F., Maurer K., Kormann R., Mucke R., Janker B. *Opt. Lasers Eng.*, **37** (2–3), 101 (2002).
4. Glownia J.H., Misewich J., Sorokin P.P. *Opt. Lett.*, **12** (1), 19 (1987).
5. Clerici M., Peccianti M., Schmidt B.E., Caspani L., Shalaby M., Giguère M., Lotti A., Couairon A., Légaré F., Ozaki T., Faccio D., Morandotti R. *Phys. Rev. Lett.*, **110**, 253901 (2013).

6. Amini-Nik S., Kraemer D., Cowan M.L., Gunaratne K., Nadesan P., Alman B.A., Miller R.J.D. *PLoS One*, **5** (9), e13053 (2010).
7. Mirov S., Moskalev I., Vasilyev S., Smolski V., Fedorov V., Martyshkin D., Peppers J., Mirov M., Dergachev A., Gapontsev V. *IEEE J. Sel. Top. Quantum Electron*., **24** (5), 1601829 (2018).
8. Steinle T., Mörz F., Steinmann A., Giessen H. *Opt. Lett*., **41** (21), 4863 (2016).
9. Woodward R.I., Hudson D.D., Fuerbach A., Jackson S.D. *Opt. Lett*., **42** (23), 4893 (2017).
10. Hanna D.C., Pointer D.J., Pratt D.J. *IEEE J. Quantum Electron*., **22** (2), 332 (1986).
11. Krylov V., Rebane A., Ollikainen O., Erni D., Wild U., Bespalov V., Staselko D. *Opt. Lett*., **21** (6), 381 (1996).
12. Didenko N.V., Losev L.L., Pazyuk V.S. *Laser Phys. Lett*., **18**, 125001 (2021).
13. Bufetov I.A., Kosolapov A.F., Pryamikov A.D., Gladyshev A.V., Kolyadin A.N., Krylov A.A., Yatsenko Y.P., Biriukov A.S. *Fibers*, **6**, 39 (2018).
14. Gladyshev A.V., Kosolapov A.F., Khudyakov M.M., Yatsenko Yu.P., Kolyadin A.N., Krylov A.A., Pryamikov A.D., Biriukov A.S., Likhachev M.E., Bufetov I.A., Dianov E.M. *Quantum Electron*., **47** (5), 491 (2017) [*Kvantovaya Elektron*., **47** (5), 491 (2017)].
15. Gladyshev A.V., Kosolapov A.F., Khudyakov M.M., Yatsenko Yu.P., Kolyadin A.N., Krylov A.A., Pryamikov A.D., Biriukov A.S., Likhachev E.M., Bufetov I.A. *IEEE J. Sel. Top. Quantum Electron*., **24**, 0903008 (2018).
16. Li H., Huang W., Cui Y., Zhou Z., Wang Z. *Opt. Express*, **28** (16), 23881 (2020).
17. Spence D.J. *Progr. Quantum Electron*., **51**, 1 (2017).
18. Konyashchenko A.V., Losev L.L., Pazyuk V.S. *Opt. Lett*., **44** (7), 1646 (2019).
19. Zhavoronkov N., Noack F., Petrov V., Kalosha V.P., Herrmann J. *Opt. Lett*., **26** (1), 47 (2001).
20. Cao L., Gao S.-F., Peng Z.-G., Wang X.-C., Wang Y., Wang P. *Opt. Express*, **26** (5), 5609 (2018).
21. Gladyshev A., Yatsenko Y., Kolyadin A., Kompanets V., Bufetov I. *Opt. Mater. Express*, **10** (12), 3081 (2020).
22. Gladyshev A.V., Astapovich M.S., Yatsenko Yu.P., Kosolapov A.F., Okhrimchuk A.G., Bufetov I.A. *Quantum Electron*., **49** (12), 1089 (2019) [*Kvantovaya Elektron*., **49** (12), 1089 (2019)].
23. Minck R.W., Terhune R.W., Rado W.G. *Appl. Phys. Lett*., **3** (10), 181 (1963).
24. Grasyuk A.Z. *Quantum Electron*., **4** (3), 269 (1974) [*Kvantovaya Elektron*., **1** (3), 485 (1974)].
25. Basiev T.T., Osiko V.V., Prokhorov A.M., Dianov E.M., in *Solid-State Mid-Infrared Laser Sources.* Ed. by I.T. Sorokina, K.L. Vodopyanov. *Topics in Applied Physics* (Berlin, Heidelberg: Springer, 2003) Vol. 89; https://doi.org/10.1007/3-540-36491-9_8.
26. Dianov E.M., in *Raman Amplifiers for Telecommunications 1*. Ed. by M.N. Islam. *Springer Series in Optical Sciences* (New York: Springer, 2004) Vol. 90/1; https://doi.org/10.1007/978-0-387-21583-9_7.
27. Leonov S.O., Voropaev V.S., Krylov A.A. *Appl. Phys. B*, **125**, 39 (2019).
28. Khudyakov M.M., Lipatov D.S., Gur'yanov A.N., Bubnov M.M., Likhachev M.E. *Opt. Lett*., **45** (7), 1782 (2020).