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Generation of radiation tunable between 350 and 600 nm and nonlinear-optical spectral transformation of femtosecond Cr : forsterite-laser pulses in submicron fused silica channels of a microstructure fibre

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***Abstract*. An efficient generation of 350 – 600-nm wavelength-tunable radiation is experimentally demonstrated through nonlinear-optical frequency conversion of femtosecond Cr : forsterite-laser pulses in an array of submicron fused silica threadlike waveguiding channels in a microstructure fibre. Nonlinear-optical spectral transformation of femtosecond pulses in such waveguide channels gives rise to new isolated spectral components with frequencies exceeding the frequency of the third harmonic of pump radiation. Some of these new spectral components are separated from the central pump frequency by a frequency gap exceeding 580 THz.**

Keywords: *ultrashort pulses, nonlinear optics, microstructure fibres.*

Frequency conversion of femtosecond laser pulses is one of the urgent problems of laser physics and quantum electronics. The standard solution to this problem is to use nonlinear-optical crystals [1]. Frequency-conversion capabilities of nonlinear crystals can be extended in certain cases by periodic poling, allowing quasi-phase matching of nonlinear optical interactions [2, 3]. The potential of photonic crystals for the frequency conversion of ultrashort pulses has been recently demonstrated, and several attractive recipes for the phase matching of nonlinear-optical processes have been suggested [4 – 6]. The main factors limiting the efficiency of frequency conversion of ultrashort pulses in nonlinear crystals and periodic structures include group-velocity dispersion and the limited spectral range of phase matching.

Microstructure [7 – 13] and tapered [14, 15] fibres possess several remarkable and unique properties, allowing highly efficient nonlinear-optical interactions to be implemented even for low-energy ultrashort laser pulses. In particular, dispersion of such fibres can be tailored by changing their geometry [16]. A high refractive index step between the core and the cladding, attainable with such fibres, provides a high degree of light confinement in the fibre core [17, 18]. Microstructure and tapered fibres offer much flexibility in phase-matching third-harmonic generation [19, 20] and four-wave mixing [21], allowing both radiation with a very broad spectrum (supercontinuum) [22] and isolated spectral components [19 – 21] to be generated with a high efficiency.

In this paper, we will demonstrate that nonlinear-optical interactions in microstructure fibres may result in highly efficient frequency conversion of femtosecond Cr : forsterite-laser pulses.

Our experiments on frequency conversion of femtosecond Cr : forsterite-laser pulses were performed with a family of fused silica microstructure fibres where the cladding, consisting of several cycles of air holes, surrounds the central fibre core with a diameter of a few micrometers (Fig. 1). Microstructure fibres were fabricated of fused silica using the technology described in detail elsewhere [23, 24]. The minimal core diameter in the fabricated family of fibres was equal to 1 μm. The air-filling fraction of the microstructure part of the cladding in the created fibres, as can be

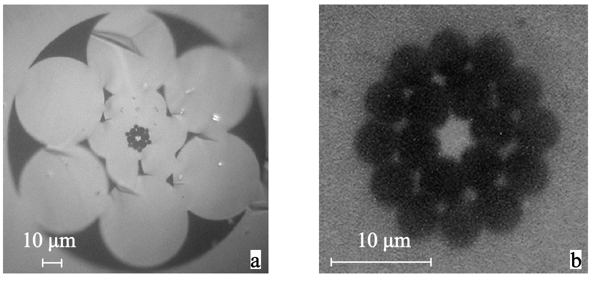

Figure 1. Cross-section images of microstructure fibres with (a) one and (b) two hexagonal cycles of air holes around the central waveguiding core.

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seen from Fig. 1, is very high, providing a high refractive index step between the core and the cladding in the fibre and strongly confining the light field in the fibre core. An array of submicron-size fused silica channels in the form of threads, bounded by the system of air holes in the fibre cladding (Fig. 1), serve as additional multiple cores of the fibre, providing a high degree of light confinement due to the high refractive-index step and allowing waveguide enhancement factors close to the physical limit, determined by the competition of diffraction and refractive-index-step waveguiding, to be achieved for nonlinear-optical processes [25].

Our experiments were performed with microstructure fibres of three types, having one, two, and three cycles of air holes around the fibre core. For each type of fibres, we examined samples with core diameters of 1, 2, 3, and 5 μm. Fibre samples were 5, 7, 10, 20, 30, and 50 cm long.

Physically, the enhancement of nonlinear-optical processes in such an array of submicron fused silica waveguiding threads is due to the same factors as the enhancement of nonlinear-optical interactions in tapered fibres. Our microstructure fibre design, in fact, combines the ideas of a holey fibre and a tapered fibre by integrating several small-core high-refractive-index-step fibres into a bundle. Such a fibre can be made as long as several hundreds meters, thus permitting length limitations, typical of tapered fibres, to be overcome by means of fibre microstructuring. The fact that the microstructure-integrated bundle includes submicron fused silica waveguiding threads with different sizes helps to achieve wavelength tuneability in the frequency conversion of ultrashort pulses (femtosecond pulses of a Cr: forsterite laser in our experiments).

The laser system employed in our experiments consisted of a Cr^{4+} : forsterite master oscillator, a stretcher, an optical isolator, a regenerative amplifier, and a compressor. The master oscillator, pumped with a fibre ytterbium laser, generated 30 – 50-fs light pulses with a repetition rate of 120 MHz. The central wavelength of this laser radiation was 1270 nm with a bandwidth of 26 nm and the mean power of about 180 mW.

Horizontally polarised 30 – 50-fs pulses were then stretched up to 700 ps in a grating stretcher (Fig. 2). Upon passing through a Faraday isolator and a $\lambda/4$ plate, the light pulses became vertically polarised. These pulses were then transmitted through a broadband polariser to be injected in the regenerative amplifier at the moment of time corresponding to maximum population inversion, created by pump pulses with a repetition rate of 1 kHz. A switch was used to set a horizontal polarisation of pulses injected into the cavity of the amplifier. An amplified pulse with an energy of 100 μJ was coupled out of the amplifier through the switch, triggered at the moment of time corresponding to optimal amplification. Radiation coming out of the amplifier was vertically polarised again. The amplified pulse was returned to the isolator along the same optical path. Radiation passing through the isolator in the backward direction experienced no change in its polarisation since polarisation rotations introduced by the $\lambda/4$ plate and the Faraday isolator compensate for each other. The pulses coupled out of the isolator through the broadband polariser were transmitted through a $\lambda/2$ plate and compressed to a 75 – 200-fs duration in a grating compressor. Approximately 50 % of pulse energy was lost at this stage.

Radiation generated by the Cr : forsterite laser system was coupled into the central core or one of the threadlike fused silica channels (Fig. 1b) in a microstructure fibre. A standard Lomo-20 objective was used (Fig. 3) to excite guided modes in the central core of the microstructure fibre. The waist length of the laser beam focused by this objective was equal to 6 – 8 μm. A Lomo-40 objective was employed to couple radiation into submicron threadlike channels of the microstructure fibre. The waist length of the focused laser beam was equal to 3 – 4 μm in this case. The duration of laser pulses at the input of microstructure fibres was about 150 fs. Spectral measurements were performed with an Ocean Optics spectrometer.

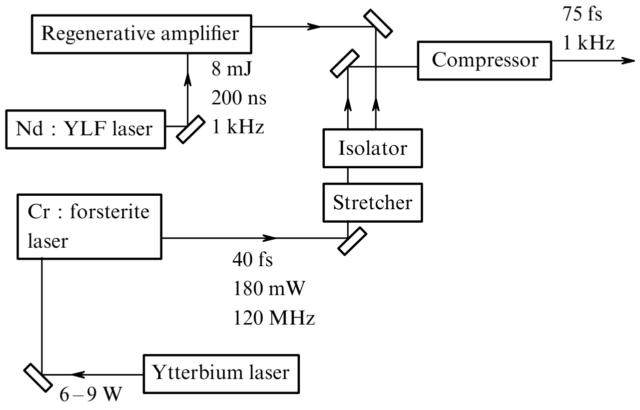

Figure 2. Femtosecond Cr : forsterite laser system with a regenerative amplifier.

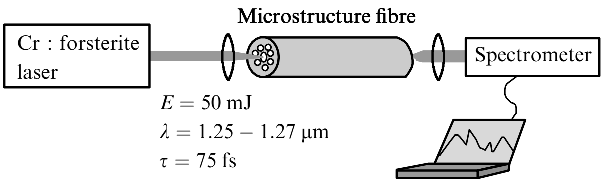

Figure 3. Diagram of the nonlinear-optical spectral transformation of femtosecond pulses in a microstructure fibre.

Femtosecond pulses experienced noticeable spectral broadening due to self-phase modulation as they propagated through the fibre (Fig. 4). With the laser pulse power being the same at the fibre input, the intensity of radiation in submicron fused silica threads was substantially (by a factor of 2.9) higher than the radiation intensity in the central core. Self-phase modulation in submicron threads was therefore much more efficient, giving rise to much broader spectra of laser pulses at the output of submicron channels (cf. Figs 4a and 4b).

Propagation of femtosecond laser pulses through microstructure fibres was accompanied by wave mixing, giving rise to new frequency components in the spectrum of radiation coming out of the fibre. Parametric four-wave mixing $2\omega_p = \omega_s + \omega_a$ (ω_p is the frequency of pump radiation and ω_s and ω_a are the frequencies of the Stokes and anti-Stokes signals, respectively) in the central fibre core resulted in the efficient generation of an anti-Stokes component centred around 530 nm with a spectral bandwidth of about 35 nm (Fig. 5). Phase matching for such

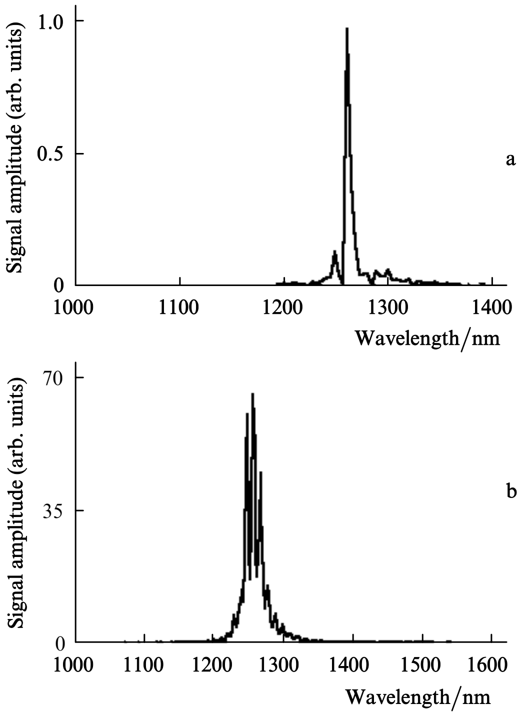

Figure 4. Spectral broadening of Cr : forsterite-laser pulses in (a) the central core of a microstructure fibre with a diameter of 3 μm and (b) a submicron threadlike waveguiding channel. The fibre length is 7 cm. The energy of laser pulses coupled into the fibre is about 50 nJ.

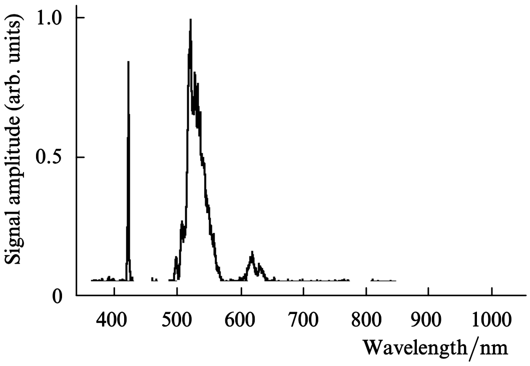

Figure 5. Spectra of the anti-Stokes signal and the third harmonic generated in the central core of a microstructure fibre with a diameter of 3 μm.

processes in microstructure fibres was analysed in earlier work [21, 26]. We also observed efficient generation of the third harmonic of pump radiation with the amplitude of the third harmonic comparable with the amplitude of the anti-Stokes signal (Fig. 5).

Nonlinear-optical spectral transformation of femtosecond Cr : forsterite-laser pulses in submicron fused silica waveguiding channels of different diameters resulted in the generation of new frequency components within a broad spectral range (Figs 6, 7). The wavelength range where the efficiency of nonlinear-optical frequency conversion reaches its maximum is determined by the dispersion properties of the waveguide channel. The size of the channel is thus the key parameter, controlling the process of nonlinear-optical frequency conversion. The possibility of tuning the frequencies of new spectral components generated by femtosecond Cr : forsterite-laser pulses in submicron fused silica channels of different diameters is illustrated in Figs 6 and 7. The spectrum of radiation presented in Fig. 6a features intense components within the range of wavelengths from 460 to 490 nm. Analysis of fibre dispersion shows that these spectral components may be produced due to parametric four-wave mixing. The spectral component appearing in the spectrum of output radiation around 420 nm (Fig. 6b) indicates efficient frequency conversion to the wavelength range close to the third harmonic of pump radiation.

An array of microstructure-integrated submicron fused silica channels implemented in our fibres provides a unique possibility of converting the frequency of femtosecond Cr : forsterite-laser pulses to the range of wavelengths

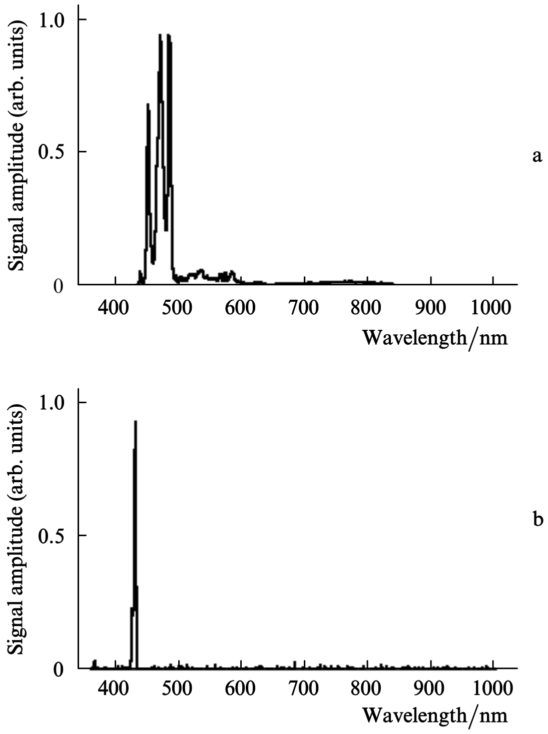

Figure 6. Spectra of (a) the anti-Stokes signal and (b) the third harmonic of pump radiation generated in submicron threadlike waveguiding channels with different diameters.

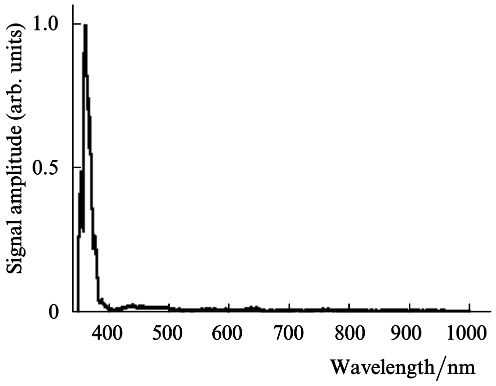

Figure 7. The spectrum of a high-frequency component with a central wavelength around 370 nm generated in a submicron threadlike waveguiding channel of a microstructure fibre. The fibre length is 7 cm. The energy of laser pulses coupled into the fibre is about 50 nJ.

shorter than the wavelength of the third harmonic. Effective cascading of nonlinear-optical processes in submicron waveguiding channels leads, as can be seen from Fig. 7, to the generation of the frequency component with a central wavelength around 370 nm and a spectral bandwidth of about 30 nm (the wavelength of the third harmonic is 420 nm for Cr : forsterite-laser radiation). This process increases the carrier frequency of femtosecond Cr : forsterite-laser pulses by more than 580 THz. The efficiency of this frequency-conversion process was as high as several percent, giving rise to blue light easily seen with a bare eye on a white screen. The developed microstructure fibre design thus allows femtosecond Cr : forsterite-laser pulses to be frequency-converted to the spectral range that is of special interest and importance for photochemical and photobiological studies, enhancing the capabilities of femtosecond Cr : forsterite lasers in femtosecond spectroscopy and time-resolved measurements, as well as extending the applicability area of such laser systems to the control of ultrafast processes in physics, chemistry, and biology.

The results of experimental studies presented in this paper demonstrate highly efficient nonlinear-optical frequency conversion of femtosecond pulses in an array of submicron fused silica threadlike channels in a microstructure fibre. The size of a waveguiding channel is the key parameter for the spectral transformation of ultrashort laser pulses. This parameter determines dispersion properties of guided modes, thus controlling the frequencies of new spectral components generated as a result of nonlinear-optical frequency conversion of ultrashort laser pulses. We have experimentally demonstrated the possibility to tune the frequencies of these new spectral components by coupling femtosecond pulses of a Cr : forsterite laser into submicron fused silica waveguiding channels with different diameters. The proposed and implemented architecture of microstructure-integrated submicron fused silica waveguiding channels is shown to offer a unique possibility of converting the frequency of femtosecond Cr : forsterite-laser pulses to the range of wavelengths shorter than the wavelength of the third harmonic, which is of special interest and importance for photochemical and photobiological applications of femtosecond pulses.

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