Cross-phase-modulation-controlled spectral transformations of ultrashort pulses in photonic-crystal fibres

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Abstract. Cross-phase modulation provides an efficient means to control spectral transformations of femtosecond laser pulses in photonic-crystal fibres with a specially designed dispersion profile. Femtosecond pulses of fundamental radiation of a Cr: forsterite laser are shown to induce an asymmetric spectral broadening of the anti-Stokes signal generated in a photonic-crystal fibre by the pulses of second-harmonic radiation of the same laser. Cross-phase-modulation-induced phase shifts control the efficiency of anti-Stokes generation in a photonic-crystal fibre, suggesting a method of switching the anti-Stokes signal on and off by varying the intensity of the control laser pulse.

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1. Introduction

Photonic-crystal fibres (PCFs) [1]–[4] find extensive applications in ultrafast nonlinear optics [5]. These fibres enhance nonlinear-optical interactions of ultrashort pulses as they provide large interaction lengths, high power densities of laser pulses in the fibre core [6], and tunable dispersion of guided modes [7]. Due to this unique combination of properties, PCFs offer attractive solutions for the creation of new sources of broadband radiation [8, 9] and frequency-tunable ultrashort pulses [4, 5] for optical metrology [10, 11], spectroscopy [12], photochemistry [13], optical signal processing [14, 15], and laser biomedicine [16, 17].

Earlier studies [18, 19] have revealed interesting properties of nonlinear-optical interactions of ultrashort laser pulses in PCFs integrating arrays of micron and submicron fused silica threadlike waveguide channels. Fibres of such an architecture provide high efficiencies of anti-Stokes frequency conversion and can be employed as multicolour sources of ultrashort pulses [5]. In this paper, we demonstrate that spectral transformations of femtosecond pulses in such PCFs can be efficiently controlled through cross-phase modulation (XPM). We will show that femtosecond pulses of fundamental radiation of a Cr : forsterite laser can induce an asymmetric spectral broadening of the anti-Stokes signal generated in a PCF by the pulses of second-harmonic radiation of the same laser. We will also discuss the possibility of using XPM to characterize ultrashort anti-Stokes pulses produced in PCFs.

2. Experimental

The laser system employed in our experiments (figure 1) consisted of a Cr$^{4+}$ : forsterite master oscillator, a stretcher, an optical isolator, a regenerative amplifier, a compressor, and a crystal for frequency doubling [20]. The master oscillator, pumped with a fibre ytterbium laser, generated 30–50 fs light pulses of radiation with a wavelength of 1.25 $\mu$m at a repetition rate of 120 MHz. These pulses were then transmitted through a stretcher and an isolator to be amplified in a Nd : YLF laser-pumped amplifier and recompressed to the 50–80 fs pulse duration with the maximum laser pulse energy up to 30 $\mu$J at 1 kHz. A 1 mm thick BBO crystal was used to generate the second harmonic of amplified Cr : forsterite laser radiation. Second-harmonic pulses, which were employed for the generation of anti-Stokes radiation, had a duration of about 300 fs and an energy of about 80 nJ.

Femtosecond pulses generated by the Cr : forsterite laser system were coupled into PCFs fabricated of fused silica using technology described in detail elsewhere [18].
Figure 1. Diagram of the experimental setup for studying nonlinear-optical spectral transformation of ultrashort laser pulses with frequency $2\omega$ (second-harmonic radiation of a Cr: forsterite laser) in photonic-crystal fibres controlled through cross-phase modulation induced by femtosecond pulses of fundamental radiation of the Cr: forsterite laser (frequency $\omega$).

Figure 2. The group index as a function of the wavelength calculated for the guided modes (1) HE$_{11}$, (2) HE$_{21}$, and (3) HE$_{31}$ in a thread-like fused silica waveguide channel having a circular cross-section with a radius of 0.6 $\mu$m. (4) Wavelength dependence of the group index for bulk fused silica. The inset presents the cross-section image of the photonic-crystal fibre. A cross-section image of a PCF employed in our experiments is shown in the inset to figure 2. An array of micron- and submicron-size fused silica channels in the form of threads, bounded by the system of air holes in the PCF cladding, serve as additional multiple cores of the fibre. These waveguide channels, as shown in [18, 19], provide high efficiencies of anti-Stokes frequency...
conversion for ultrashort laser pulses. The mismatch of the group velocities of the pump pulse and the anti-Stokes signal limits the efficiency of this process and leads to an increase in the duration of the anti-Stokes signal relative to the pump pulse. Special PCFs were produced for the purposes of this work, with a dispersion profile (figures 2 and 3) reducing the group-velocity mismatch for the pump pulse and the anti-Stokes signal centred at a wavelength of 414 nm.

A LOMO-40 objective was employed to couple pump radiation into thread-like fused silica waveguide channels in the PCF. The confocal parameter of the laser beam focused with this objective was equal to 6–8 µm. Propagation of second-harmonic pulses of Cr:forsterite laser radiation through PCFs was accompanied by wave mixing, giving rise to new frequency components in the spectrum of radiation coming out of the fibre. These spectral components were detected with an Ocean Optics spectrometer, placed behind the PCF. Parametric four-wave mixing \(2\omega_p = \omega_S + \omega_a\) (\(\omega_p\) is the frequency of pump radiation and \(\omega_S\) and \(\omega_a\) are the frequencies of the Stokes and anti-Stokes signals, respectively) in one of the waveguide channels of the PCF resulted in the efficient generation of anti-Stokes radiation with a central wavelength of 414 nm (figure 4). Phase matching for such processes in microstructure fibres was analysed in earlier work [21]–[23].

3. Results and discussion

Femtosecond pulses of fundamental radiation of the Cr:forsterite laser were coupled into the PCF with a delay time \(\tau\) with respect to second-harmonic pulses. Ultrashort pulses of fundamental radiation were used to control the spectrum of the anti-Stokes signal generated by second-harmonic pulses. With the second harmonic blocked, control pulses of fundamental radiation resulted in no emission within the range of wavelengths from 400 to 430 nm. An anti-Stokes signal centred at 414 nm was detected only when the second harmonic was switched on. Intensity distribution of the anti-Stokes signal in the cross-section of the PCF indicates that this signal

\[D, \text{ps/(nm km)} \]

\[\lambda, \mu\text{m}\]

**Figure 3.** Group-velocity dispersion \(D\) as a function of the wavelength calculated for the guided modes: 1, HE\(_{11}\); 2, HE\(_{21}\); and 3, HE\(_{31}\) in a thread-like fused-silica waveguide channel having a circular cross-section with a radius of 0.6 µm.
is generated as a mixture of guided modes. This finding agrees well with the predictions of theoretical analysis [19, 23], which suggests the possibility of phase-matched four-wave mixing $2\omega_p = \omega_{St} + \omega_a$ with the anti-Stokes signal emitted in higher-order guided modes.

For small delay times $\tau$, the control pulse of fundamental radiation gives rise to a considerable spectral broadening of the anti-Stokes signal (figure 4), related to cross-phase modulation. The nonlinear phase shift induced by XPM is given by [24, 25]

$$\Phi_{XPM}(\eta, z) \propto \chi^{(3)}(\omega_s; \omega_s, \omega_p, -\omega_p) \int_0^z |A_p(\eta - \xi/\sigma, 0)|^2 d\xi,$$

where field $\omega_p$ is the frequency of pump radiation; $\omega_s$ is the frequency of the probe pulse; $\chi^{(3)}(\omega_s; \omega_s, \omega_p, -\omega_p)$ is the third-order nonlinear-optical susceptibility; $A_p(\xi, 0)$ is the initial envelope of the pump pulse, $1/\sigma = 1/u_1 - 1/u_2$; and $u_1$ and $u_2$ are the group velocities of the pump and probe pulses, respectively. The corresponding spectral broadening can be written as

$$\delta\omega_{XPM}(\eta, z) \propto \chi^{(3)}(\omega_s; \omega_s, \omega_p, -\omega_p)\sigma[|A_p(\eta, 0)|^2 - |A_p(\eta - z/\sigma, 0)|^2].$$

In our experiments, XPM results in an asymmetric spectral broadening of the anti-Stokes signal, which displays a predominant red shift (figure 4). Such a character of spectral broadening can be explained by group-delay effects in the regime of anomalous dispersion [25]–[27]. For HE$_{11}$ guided modes of thread-like fused silica waveguide channels, the group velocity of the control pulse (in contrast to a bulk sample of fused silica, curve 4 in figure 2) is lower than the group velocity of the anti-Stokes signal. Cross-phase modulation then predominantly occurs on the leading edge of the control pulse, leading to the blue shifting of the anti-Stokes signal.
Cross-phase modulation observed in our experiments can be employed for the metrology of the anti-Stokes signal generated in a PCF. The control pulse of fundamental radiation has a considerable influence on the spectrum of the anti-Stokes signal within the time interval $\Delta \tau \approx 700 \text{ fs}$. Since the duration of the pulse of fundamental radiation is much shorter than $\Delta \tau$, this result gives an upper-bound estimate for the duration of the anti-Stokes pulse: $\tau_a < 700 \text{ fs}$.

A more precise measurement of the duration of this pulse can be performed in the regime where the group velocities of the anti-Stokes and control pulses are matched.

Generation of subpicosecond pulses of anti-Stokes radiation becomes possible in our experiments due to a special dispersion profile of guided modes, reducing the group-velocity mismatch between the second-harmonic pulse and the anti-Stokes signal. Figure 2 presents the dependences of group indices $n_{g\nu} = c/v_{g\nu}(v_{g\nu} = (\partial \beta_{\nu}/\partial \omega)^{-1}$ is the group velocity, $\beta_{\nu}$ is the propagation constant, and $c$ is the speed of light in vacuum) on the wavelength for guided modes $\text{HE}_{\nu,1}$ of a thread-like fused silica waveguide channel having a circular cross-section with a radius of $0.6 \mu \text{m}$. For comparison, dotted curve 4 in figure 2 displays the group index $n_g = n[1 + (\omega/n)(\partial n/\partial \omega)]$ (n is the refractive index) calculated as a function of the wavelength for a bulk sample of fused silica. Group-velocity dispersion $D$ calculated as a function of the wavelength for the waveguide modes $\text{HE}_{2,1}$ is presented in figure 3. Analysis of dispersion properties of guided modes in thread-like fused silica channels of our PCFs shows that the dispersion length for pump pulses with a duration of about 300 fs is approximately equal to 2 m. This result agrees well with direct cross-correlation measurements, which indicate that the dispersion spreading of the pump pulse under conditions of our experiments is negligible. Group-velocity mismatch gives rise to a spatial walk-off of the pump pulse and the anti-Stokes signal. With a PCF length equal to $L = 7 \text{ cm}$, the group-delay time, $t_{gw} = L|n_{g\nu}(\omega_p) - n_{g\nu}(\omega_s)|/c$, for the $\text{HE}_{2,1}$ mode is approximately equal to 700 fs. Thus, PCFs with a special dispersion profile designed for our XPM experiments allow the group delay of femtosecond pump pulses and the nonlinear signal within the nonlinear interaction length to be reduced by more than an order of magnitude relative to a bulk sample of fused silica, where $t_w = L|n_g(\omega_p) - n_g(\omega_s)|/c \approx 8.3 \text{ ps}$.

The pump-intensity-dependent phase shift induced by XPM at the frequencies of the second harmonic and the anti-Stokes signal suggests a way to control the phase matching for the PCF modes involved in the parametric four-wave mixing process. This effect can be employed to switch the anti-Stokes signal on and off by varying the intensity of the pump field. To demonstrate this anti-Stokes switching performance of a PCF, 100 fs pulses of fundamental Cr:forsterite laser radiation and 300 fs pulses of its second harmonic were coupled into the central core of a 7 cm fused silica PCF with the cross-section structure shown in the inset of figure 5. The diameter of the central core in this PCF was 2.8 $\mu \text{m}$. As before, second-harmonic pulses serve as a pump field to generate the anti-Stokes signal through the $2\omega_p = \omega_{\text{St}} + \omega_a$ FWM process. The anti-Stokes signal is centred at 407 nm in this case. Femtosecond pulses of the fundamental radiation are used as a control field to switch the anti-Stokes signal on and off. Figure 5 displays the spectra of the second-harmonic pulse measured at the output of the PCF for two different delay times $\tau$ between the control and pump pulses. In figure 6, we present the spectra of anti-Stokes emission measured within the range of wavelengths from 400 to 450 nm for the values of the delay time. With $\tau = 600 \text{ fs}$ (dashed navy lines in figures 5 and 6), the control pulse has virtually no influence on the FWM process, with the spectrum of the second-harmonic pump pulse and the spectrum of the anti-Stokes signal at the output of the PCF being identical to the pump and anti-Stokes spectra measured with no control pulse at the input of the PCF. As $\tau$ decreases, however, the second-harmonic pulse transmitted through the PCF displays noticeable
Figure 5. The spectra of the second-harmonic pulse measured at the output of a 7 cm PCF for two different delay times between this pulse and the control pulse of 1.25 µm Cr : forsterite laser radiation: τ = 600 fs (dashed navy line) and 0 (solid olive line). The energy of the control pulse is 0.1 µJ. The inset shows the image of a PCF with a central core diameter of 2.8 µm.

Figure 6. The spectra of anti-Stokes emission at the output of a 7 cm PCF for two different delay times between the control pulse of 1.25 µm Cr : forsterite laser radiation and the second-harmonic pulse used as a pump: τ = 600 fs (dashed navy line) and 0 (solid olive line).

pump-intensity-dependent spectral distortions related to the XPM effect (the solid olive line in figure 5). Anti-Stokes signal generation is almost totally suppressed in this regime (the solid olive line in figure 6) due to the XPM-induced phase mismatch between the pump, Stokes and anti-Stokes modes of the PCF involved in the FWM process. This effect of XPM-controlled anti-Stokes generation in a PCF demonstrates the possibility of building PCF switching devices, logic gates and photonic information-processing components.

4. Conclusion

Experiments presented in this paper demonstrate that spectral transformations of femtosecond pulses in photonic-crystal fibres can be controlled through cross-phase modulation. Femtosecond
pulses of fundamental radiation of a Cr: forsterite laser have been shown to induce an asymmetric spectral broadening of the anti-Stokes signal generated in a photonic-crystal fibre by the pulses of second-harmonic radiation of the same laser. We have demonstrated that cross-phase-modulation-induced phase shifts control the efficiency of anti-Stokes generation in a photonic-crystal fibre, suggesting the method of switching the anti-Stokes signal on and off by varying the intensity of the control laser pulse, thus offering a promising strategy for the creation of PCF switching devices, logic gates, and information processing components.

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