Soft-glass photonic-crystal fibers for frequency shifting and white-light spectral superbroadening of femtosecond Cr:forsterite laser pulses

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Structural dispersion and nonlinearity management of multicomponent-glass photonic-crystal fibers is shown to allow wavelength-tunable frequency shifting and white-light spectral transformation of femtosecond Cr:forsterite laser pulses. Launching 200 fs pulses of 1.25 μm Cr:forsterite laser radiation into such dispersion-managed soft-glass photonic-crystal fibers in the regime of anomalous dispersion, we demonstrate spectrally tailored supercontinuum generation and frequency upshifting, yielding isolated spectral components with central wavelengths ranging from 400 to 900 nm. © 2006 Optical Society of America

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1. INTRODUCTION

Photonic-crystal fibers (PCFs)^{1,2} have proved to be a powerful tool of femtosecond laser technologies, leading to a tremendous enhancement of the capabilities of laser sources of ultrashort pulses and few-cycle field waveforms. Dispersion tunability³ and strong field confinement in the fiber core in PCFs^{4–6} are the key factors providing remarkably high efficiencies of nonlinear-optical phenomena, thus allowing the creation of novel fiber-optic broadband light sources⁷ and efficient frequency shifters.⁸ Such PCF components are currently extensively used in many areas of ultrafast science, giving an excess to the carrier-envelope phase,^{9,10} allowing the generation of stabilized frequency combs for optical metrology and femtosecond clockwork,^{11–13} permitting creation of novel optical coherence tomographs¹⁴ and generators of correlated photons,¹⁵ simplifying pump-seed synchronization in few-cycle optical parametric chirped-pulse amplifiers,¹⁶ improving the performance of nonlinear spectrographs¹⁷ and microscopes¹⁸ based on coherent anti-Stokes Raman scattering, and expanding the applicability range of femtosecond laser sources to photochemistry and photobiology.¹⁹

Several attractive PCF-based strategies have been demonstrated for highly efficient nonlinear-optical spectral transformation of femtosecond Ti:sapphire laser pulses. These solutions are based on dispersive-wave emission by solitons,²⁰ four-wave mixing,^{21,22} modulation instabilities,²³ stimulated Raman scattering,²¹ and soliton self-frequency shift.²⁴ Extension of these strategies of frequency conversion to other input wavelengths and other types of short-pulse sources requires careful PCF dispersion management and sometimes necessitates optimization of the fiber material. Several types of nonsilica PCFs have been used for the spectral transformation of laser radiation in the near-infrared range. An extruded soft-glass SF6 PCF has been used²⁵ for ultrabroad supercontinuum generation seeded by 100 fs pulses of 1550 nm radiation from an optical parametric amplifier. An SF6 PCF was later shown²⁶ to allow supercontinuum generation with 200 pJ pulses of 1560 nm radiation from an all-fiber diode-pumped erbium-fiber oscillator–amplifier system used as a pump source. Soliton self-frequency shifts²⁷ of mode-locked erbium-fiber laser operating at 1556 nm have been demonstrated with lead-silicate PCFs.²⁸ Phase-

matched third-harmonic generation by femtosecond Cr:forsterite laser pulses has been observed in multicomponent-glass PCFs.²⁹

We will show in this work that nonsilica PCFs can offer interesting alternatives to silica microstructure fibers for supercontinuum spectral transformation and frequency upshifting of femtosecond pulses within the 1.2–1.3 μm range of central wavelengths. This range of wavelengths, covered by femtosecond Cr:forsterite lasers, is of special importance for biomedical imaging.^{30–32} As compared with 800 nm Ti:sapphire laser radiation, femtosecond pulses of 1.2–1.3 μm radiation can provide a significant improvement in penetration depth for rapidly growing nonlinear-optical bioimaging techniques applied to human skin tissues and other biological objects. Already noticeable for the fundamental wavelengths of the Ti:sapphire and Cr:forsterite lasers (800 nm versus 1.25 μm), this improvement becomes quite dramatic for the second and third harmonics of laser radiation, making a Cr:forsterite laser an advantageous source for bioimaging schemes based on second- and third-harmonic generation, as well as on coherent anti-Stokes Raman scattering. Fiber-optic components for the frequency conversion of Cr:forsterite laser pulses would thus allow the creation of compact devices for the wavelength-selective imaging of biotissues. We will show in this work that the structural dispersion and nonlinearity management of multicomponent-glass PCFs enables wavelength-tunable frequency shifting and white-light spectral transformation of femtosecond Cr:forsterite laser pulses. By coupling 200 fs pulses of 1.24 μm Cr:forsterite laser radiation into different types of multicomponent-glass PCFs, where the zero group-velocity dispersion (GVD) wavelength is tuned by scaling the sizes of the fiber structure, we demonstrate spectrally tailored supercontinuum generation and frequency upshifting, yielding isolated spectral components with central wavelengths ranging from 400 to 900 nm.

2. DISPERSION AND NONLINEARITY OF SOFT-GLASS PHOTONIC-CRYSTAL FIBERS

Experiments were performed with two types of multicomponent-glass PCFs, fabricated with the use of the standard PCF technology.^{1,2} PCFs of both types were made of multicomponent glass containing approximately 90% of S93-1 glass (55% of SiO_2 , 30% of PbO , 2.0% of Al_2O_3 , 3.8% of Na_2O , and 9.2% of K_2O) with additions of S95-2 glass (69.6% of SiO_2 , 4.0% of Al_2O_3 , 2.8% of B_2O_3 , 6.9% of CaO , 9.0% of Na_2O , and 7.7% of K_2O). In PCFs of the first type (Fig. 1, top), the solid fiber core is surrounded by a high-air-filling-fraction Kagome-lattice-type cladding. In PCFs of the second type (Fig. 1, bottom), the solid fiber core is surrounded by a single ring of thin-wall capillaries whose outer diameters are equal to the diameter of the PCF core. The outer part of the microstructure cladding consists of 11 rings of capillaries with outer diameters approximately three times larger than the diameter of the PCF core and high air-filling fraction.

Dispersion and nonlinearity of the PCFs of both types were managed by scaling the geometric sizes of the PCF structure. Technologically, this was realized by using the same preform to fabricate PCFs with the same type of the

Fig. 1. SEM images of soft-glass photonic-crystal fibers. The p axis shows polarization of the input field.

structure, but with different magnifying factors. This procedure allowed us to scale the sizes of PCF structure without changing its geometry. In Fig. 2, we plot the GVD as a function of the wavelength calculated for the PCF of the first type using the finite-element method, adapted to include the specific fiber structure and material dispersion. As the PCF core diameter is changed from 1.5 to 4 μm , the zero-GVD wavelength is tuned from 814 to 1175 nm (curves 1–4 in Fig. 2). For comparison, the material GVD of the multicomponent glass employed to fabricate PCFs is presented by line 5 in Fig. 2. The material part of the GVD passes through zero at 1.66 μm .

PCFs with smaller cores provide a stronger confinement of guided modes to the fiber core, leading to higher nonlinearity. To quantify this nonlinearity enhancement for the fibers used in this work, we consider the nonlinear coefficient, defined as $\gamma = (2\pi/\lambda) \times [\int \int n_2(x,y) S_z^2 dx dy / |\int \int S_z dx dy|^2]$, where λ is the wavelength, $n_2(x,y) = n_2 \xi(x,y)$, n_2 is the nonlinear refractive index of the fiber material, $\xi(x,y)$ is the two-dimensional spatial profile of the nonlinear refractive index in the PCF cross section, and S_z is the z component of the Poynting vector (with the z axis chosen in the direction of waveguiding). In Fig. 2 (top), this nonlinear coefficient is

plotted as a function of the wavelength λ for the PCFs of the first type. More compact modes of PCFs with a smaller core diameter, as can be seen from these plots, are characterized by higher nonlinear coefficients γ than modes with larger core diameters. As the radiation wavelength increases, on the other hand, the nonlinearity lowers because of the explicit dependence of γ on λ and because of the increase in the effective mode area with growing λ .

To compare dispersion and nonlinear properties of our multicomponent-glass PCFs with the properties of fused-silica fibers, we also plot in Fig. 2 (bottom, curves 6 and 7) the GVD and the nonlinear coefficient for fibers consisting of a circular fused-silica core and an air cladding with core diameters equal to those of first-type soft-glass PCFs employed in our experiments. With the nonlinear refractive index of our soft-glass PCF, $n_2 \approx 2.5 \times 10^{-16} \text{ cm}^2/\text{W}$, being very close to the n_2 of fused silica, air-cladded fused-silica rods provide nonlinear coefficients γ close to those of

soft-glass PCFs with equal core diameters (see Fig. 2, top). However, dispersion properties of silica fibers, as can be seen from Fig. 2, drastically differ from those of soft-glass PCFs. While the GVD of soft-glass PCFs passes through zero at 910 and 980 nm for core diameters 2 and 2.4 μm (curves 2 and 3), respectively, zero GVD for silica rods with the same diameters is achieved at 715 and 767 nm (curves 6 and 7), respectively. Thus, owing to a different material dispersion (material GVD for silica is shown by curve 8 in Fig. 2, bottom), zero-GVD wavelengths of silica fibers lie much further away from the central wavelength of Cr:forsterite laser radiation (1.25 μm in our experiments) than in the case of the soft-glass PCFs with nearly identical nonlinearities. This comparison suggests that the soft-glass PCFs with the above-specified parameters are better suited for white-light spectral broadening of Cr:forsterite laser pulses. In particular, dispersion of soft-glass PCFs allows phase matching for Cherenkov-type emission of dispersive waves^{33,34} in the visible by solitons produced by Cr:forsterite laser pulses. Such phase matching is illustrated by the upper curves in Figs. 3(a) and 3(b), showing the mismatch $\delta = \beta_s(\lambda_0) - \beta(\lambda_d)$ of the propagation constant $\beta(\lambda_d)$ of dispersive-wave emission at the wavelength λ_d in the fundamental mode of the fiber and the propagation constant of the soliton $\beta_s(\lambda_0)$ centered at the pump wavelength $\lambda_0 = 1.25 \mu\text{m}$. This resonant emission of dispersive waves, as shown by earlier studies (e.g., Ref. 20) and as confirmed by the results of experiments presented below in this paper, is an important ingredient of supercontinuum generation in PCFs.

Although the nonlinear refractive index n_2 in our PCFs remained close to that of fused silica, higher n_2 values can be found in other fiber-technology-compatible materials. Excellent reviews of high- n_2 glasses and high-Raman-gain materials, offering new interesting possibilities in nonlinear fiber optics, have been provided by Friberg and Smith³⁵ and Stegeman *et al.*³⁶ As the fiber material is changed, the material dispersion profile is modified as well. In particular, Petropoulos *et al.*²⁸ demonstrated PCFs made of high- n_2 lead-silicate SF57 glass. The nonlinear refractive index for this glass, $n_2 \approx 4.1 \times 10^{-15} \text{ cm}^2/\text{W}$, is more than 1 order of magnitude higher than that for fused silica. The zero-GVD wavelength of bulk SF57 glass is $\sim 1970 \text{ nm}$, i.e., is substantially shifted toward longer wavelength with respect to the zero-GVD wavelength of bulk silica. Zero-GVD wavelengths for PCF modes also tend to be redshifted relative to zero-GVD for identical PCF structures, but made of fused silica. For chalcogenide glasses, the increase in material nonlinearity is even more dramatic, as n_2 may be as high as $1.1 \times 10^{-13} \text{ cm}^2/\text{W}$ (Ref. 37). In addition to their high nonlinearity and Raman gain, chalcogenide glasses can be transparent for wavelengths up to about 5 μm , showing promise³⁸ for the development of fiber-optic switches and sensors, as well as Raman fiber lasers and amplifiers in the near-, mid-, and long-wave IR spectral regions. Fabrication of chalcogenide PCFs was reported by Monro *et al.*³⁹

The laser system used in our experiments³² consisted of a Cr⁴⁺:forsterite master oscillator, a stretcher, an optical isolator, a regenerative amplifier, and a compressor. The

Fig. 2. Bottom, GVD as a function of the wavelength for soft-glass PCFs with the cross-section structure shown in Fig. 1 (top) and a core diameter of (1) 1.5, (2) 2, (3) 2.4, and (4) 4 μm . Material GVD for PCF glass is shown by curve 5. Also shown is the GVD for a fiber consisting of a circular fused-silica core and an air cladding with a core diameter of (crosses, curve 6) 2 μm and (circles, curve 7) 2.4 μm . Material GVD for silica is shown by curve 8 (triangles). Top, the nonlinear coefficient γ as a function of the wavelengths for the same type of PCF with a core diameter of (1) 2 and (2) 2.4 μm , compared with the nonlinear coefficients for circular fused-silica fibers with an air cladding and a core diameter of (crosses) 2 μm and (circles) 2.4 μm .

Fig. 3. Output spectra of a soft-glass photonic-crystal fiber with a cross-section structure shown in Fig. 1 (top) and a core diameter of (a) 2.4 and (b) 2 μm . The input amplified Cr:forsterite laser pulse has a pulse width of 200 fs and an energy of (a) (1) 10, (2) 16, (3) 25, and (4) 40 nJ and (b) (1) 20, (2) 30, and (3) 40 nJ. The fiber length is 12 cm. Curve 5 in Fig. 3(a) and curve 4 in Fig. 3(b) show the propagation-constant mismatch δ between the soliton centered at 1.25 μm and dispersive waves guided by the fundamental mode of the PCF as a function of the wavelength of the dispersive wave. The horizontal dashed line in Fig. 3(b) represents the average maximum level formally defined as the half-sum of the amplitudes of the peaks centered at 580 and 860 nm; $\delta\omega_{20\%}$ is the bandwidth of the output radiation measured at the level of 20% of the average maximum. (c) The entire radiation spectrum measured at the output of the first-type PCF with a core diameter of 2.0 μm and a length of 15 cm. The energy of the input pulse is 32 nJ; $\delta f_{20\text{dB}}$ is the width of the total output spectrum measured at the level of 20 dB of the maximum spectral intensity.

master oscillator, pumped with a fiber ytterbium laser, generated 30–60 fs light pulses of radiation with a wavelength of 1.23–1.25 μm 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 200 fs pulse duration with the maximum laser pulse energy up to 20 μJ at 1 kHz. The energy of laser pulses used in experiments presented in this paper ranged from 0.5 up to 200 nJ. Because of the slight form anisotropy of the PCF, output radiation spectra were sensitive to the polariza-

tion of the input field. Below in this paper, we present experimental results obtained with an input field linearly polarized along the p axis, shown in Fig. 1.

3. RESULTS AND DISCUSSION

For both first- and second-type PCFs used in our experiments, the central wavelength of Cr:forsterite laser pulses falls within the range of anomalous dispersion. Such pulses can therefore form solitons in the fiber. High-order dispersion induces wave-matching resonances between solitons and dispersive waves,^{33,34} giving rise to intense blueshifted emission, observed as prominent features in the PCF output spectra [curves 1–4 in Fig. 3(a) and curves 1–3 in Fig. 3(b)]. The central wavelength of this blueshifted emission correlates well with the wavelength where dispersive waves guided in the fundamental mode of the PCF are phase matched with the soliton produced by Cr:forsterite laser pulses. In particular, our analysis for the first-type PCF with a core diameter of 2.4 μm predicts soliton dispersive-wave phase matching around 613 nm [curve 5 in Fig. 3(a)]. This is exactly where the spectrum of the blueshifted signal is centered for amplified input pulses with energies of 8–12 nJ transmitted through the first-type PCF with a core diameter of 2.4 μm and a length of 12 cm. As the input energy is increased, this component tends to broaden and its high-frequency edge becomes shifted toward shorter wavelengths, owing to the generation of multiple solitons and cross-phase modulation (the influence of cross-phase modulation on supercontinuum generation in PCFs was recently highlighted by Genty *et al.*⁴⁰), eventually giving rise to a broadband emission at the output of the PCF.

First-type PCFs with a core diameter of 2.0 μm , as can be seen from Fig. 2 (top), provide a higher nonlinearity than PCFs with a core diameter of 2.4 μm do, leading to the generation of broader output spectra for the same input energy and fiber length. Similar to 2.4 μm core-diameter PCFs, the dominant peak in the output spectra of PCFs with a core diameter of 2.0 μm is observed at the wavelength where a dispersive-wave guided mode of the PCF is phase matched with the soliton pump field [as illustrated by curve 4 in Fig. 3(b)]. This resonant dispersive-wave emission gives rise to a peak at 530–540 nm in Fig. 3(b). To quantify spectral broadening, we define the average maximum level of spectral intensity for the output spectra with the largest spectral extension [curve 3 in Fig. 3(b)] as the half-sum of the amplitudes of second most intense peaks in the output spectrum, observed at 580 and 860 nm. This formal definition of the maximum level [shown by the horizontal dashed line in Fig. 3(b)] does not include the most intense, but narrowband dispersive-wave peak at 530 nm. The bandwidth $\delta\omega$ of the output radiation is then measured at the level of 20% of the average maximum, giving, as shown in Fig. 3, $\delta f_{20\%} = \delta\omega_{20\%}/2\pi \approx 320$ THz, with the intensity spectrum stretching from 480 to 970 nm for an input pulse energy of 40 nJ. The maximum energy of radiation emitted in the 480–970 nm spectral range under these conditions is 70 pJ. In view of 10% in-coupling efficiency, this corresponds to a frequency-conversion efficiency of approximately 2%.

Figure 3(c) displays the entire radiation spectrum measured at the output of the first-type PCF with a core diameter of $2.0\ \mu\text{m}$ and a length of 15 cm. Defining the total width $\delta f_{20\text{ dB}}$ of the output PCF spectrum at the level of 20 dB below the maximum, we find from Fig. 3(c) that $\delta f_{20\text{ dB}} \approx 550\ \text{THz}$. The output spectrum is dominated by two broad radiation bands, corresponding to the visible emission (from approximately 500 to 930 nm) and the spectrally broadened pump (from 1100 to 1320 nm). These two bands are separated by a dip in the output spectrum, stretching from approximately 930 to 1100 nm. This dip tends to decrease with the increase in the fiber length, as the spectral widths of both the visible and infrared parts of the output spectrum continue to grow. However, the length of PCF samples in our experiments was limited by fiber losses, estimated as 2–10 dB/m. With the dominant loss mechanisms in our conditions originating from an incomplete control of glass impurities, an improved fabrication procedure should allow the level of losses for multicomponent-glass PCFs to be reduced to several tens of decibels per kilometer. Although such a level of optical losses will still be substantially higher than the minimum loss levels below decibel per kilometer attainable with fused-silica PCFs,¹ efficient frequency upconversion provided by multicomponent-glass PCFs presented here, resulting in the generation of intense spectral components in the visible, offers attractive solutions for coherent nonlinear spectroscopy¹⁷ and microscopy,¹⁸ as well as for time-resolved spectroscopic measurements.⁴¹ Because of PCF losses and the low incoupling efficiency (about 10%), high-input powers were needed in our measurements, requiring the use of a Cr:forsterite amplifier, for efficient spectral transformations of Cr:forsterite laser pulses. Even with these problems remaining to be resolved, multicomponent-glass PCFs can enhance spectral transformations of Cr:forsterite laser pulses relative to silica PCFs, as the former fibers can simultaneously provide a suitable dispersion regime (Fig. 2) and a small effective mode area, leading to a high nonlinearity for the central wavelength of Cr:forsterite laser radiation (the inset in Fig. 2).

The phase matching between the soliton produced by the input laser pulse and the dispersive wave, which defines the frequency of the dominant peaks in output spectra of PCFs, is controlled by the dispersion of the fiber. The central frequency of the blueshifted signal in the output of PCFs can thus be adjusted by modifying the dispersion profile of the fiber. This option is demonstrated by the results of our experiments performed with second-type PCFs. The GVD profiles of waveguide modes were modified by scaling the geometric sizes of the fiber without changing the type of the structure shown in Fig. 1 bottom. The core diameter of PCFs fabricated for these experiments was varied from 0.9 to $3.8\ \mu\text{m}$. Figure 4 shows the spectra of the blueshifted output of the fiber, adjusted by changing the fiber to vary the offset $\Delta = \lambda_0 - \lambda_z$ between the central wavelength of the input laser field, λ_0 , and the zero-GVD wavelength λ_z . With the PCF length remaining unchanged (10 cm), larger blueshifts are achieved by increasing the offset Δ (see curves 1–6 in Fig. 4). The power of the input laser field needs to be increased for larger Δ in these experiments to keep the amplitude of the blue-

Fig. 4. Intensity spectra of the dispersion-managed blueshifted output of soft-glass PCFs. The wavelength offset δ is 50 nm (1), 110 (2), 150 (3), 190 (4), 220 (5), and 300 nm (6).

shifted signal constant. In these experiments, dispersion-managed soft-glass PCFs are shown to serve as frequency shifters of femtosecond Cr:forsterite laser pulses, providing an anti-Stokes output adjustable across the range of wavelengths from 400 to 900 nm.

4. CONCLUSION

We have shown in this work that the structural dispersion and nonlinearity management of multicomponent-glass PCFs allows a wavelength-tunable frequency shifting and white-light spectral transformation of femtosecond Cr:forsterite laser pulses. We experimentally demonstrated frequency shifting and white-light spectral transformation of femtosecond Cr:forsterite laser pulses in nonsilica PCFs. This study allowed us to identify important advantages of multicomponent-glass PCFs over silica microstructure fibers for the spectral transformation of laser pulses in the $1.2\text{--}1.3\ \mu\text{m}$ spectral range. By coupling 200 fs pulses of $1.24\ \mu\text{m}$ Cr:forsterite laser radiation into different types of multicomponent-glass PCFs, where the zero-GVD wavelength is tuned by scaling the sizes of the fiber structure, we have demonstrated spectrally tailored supercontinuum generation and frequency upshifting, yielding isolated spectral components with central wavelengths ranging from 400 to 900 nm.

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