Highly nonlinear photonic-crystal fibers for the spectral transformation of Cr: forsterite laser pulses

A.B. Fedotov^{a,b}, E.E. Serebryannikov^a, A.A. Ivanov^{b,c}, D.A. Sidorov-Biryukov^b,
L.A. Melnikov^d, A.V. Shcherbakov^e, Chi-Kuang Sun^f, M.V. Alfimov^c, A.M. Zheltikov^{a,b,*}

^a Physics Department, Faculty of Physics, M.V. Lomonosov Moscow State University, Vorob'evy gory, Moscow 119992, Russian Federation

^b International Laser Center, M.V. Lomonosov Moscow State University, Vorob'evy gory, Moscow 119992, Russian Federation

^c Center of Photochemistry, Russian Academy of Sciences, ul. Novatorov 7a, Moscow 117421, Russian Federation

^d N.G. Chernyshevsky Saratov State University, ul. Moskovskaya 155, 410026 Saratov, Russian Federation

^e Institute of Glass Structure Technology and Equipment, pr. Stroitelei 1, 410044 Saratov, Russian Federation

^f National Taiwan University, Institute of Electro-Optical Engineering, Taipei 10617, Taiwan

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Abstract

We demonstrate novel photonic-crystal fibers (PCFs) fabricated of a highly nonlinear glass. Dispersion profiles and nonlinearity of these fibers are tailored with an array of submicron holes in the fiber core. With the PCF structure designed to provide a nonlinearity on the order of $10^3 \text{ W}^{-1} \text{ km}^{-1}$ at the radiation wavelength of $1 \mu\text{m}$ and a fundamental-mode dispersion profile with zero group-velocity dispersion around $1.19 \mu\text{m}$, unamplified femtosecond Cr: forsterite laser pulses are efficiently frequency-converted into the 540–1000-nm wavelength range through solitonic spectral-transformation mechanisms and four-wave mixing.

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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 tunability [3] 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 sources [7] 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 tomographs [14] and generators of correlated photons [15], simplifying pump-seed synchronization in few-cycle optical parametric chirped-pulse amplifiers [16], improving the performance of nonlinear spectrographs [17] and microscopes [18] based on coherent anti-Stokes Raman scattering, and expanding the applicability range of femtosecond laser sources to photochemistry and photobiology [19,20].

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 [21], four-wave mixing [22], modulation instabilities [23], stimulated Raman scattering [22], and soliton

* Corresponding author. Address: Physics Department, Faculty of Physics, M.V. Lomonosov Moscow State University, Vorob'evy gory, Moscow 119992, Russian Federation. Tel.: +7 095 939 5174; fax: +7 095 939 3113.

E-mail address: zheltikov@phys.msu.ru (A.M. Zheltikov).

self-frequency shift [24]. Extension of these strategies of frequency conversion to other input wavelengths and other types of short-pulse laser sources requires careful PCF dispersion management and sometimes necessitates optimization of fiber material [25]. Several types of non-silica PCFs have been earlier used for the spectral transformation of laser radiation in the near-infrared range. An extruded soft-glass SF₆ PCF has been used [26] for ultrabroad supercontinuum generation seeded by 100-fs pulses of 1550-nm radiation from an optical parametric amplifier. An SF₆ PCF has been later shown [27] 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 shift of mode-locked erbium fiber laser operating at 1556 nm has 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]. In a recent paper, Gopinath et al. [30] have reported on highly nonlinear bismuth-oxide fibers for supercontinuum generation and femtosecond pulse compression. Excellent reviews of high- n_2 glasses and high-Raman-gain fiber-technology-compatible materials have been provided by Friberg and Smith [31] and Stegeman et al. [32].

In this work, we demonstrate novel PCFs fabricated of a highly nonlinear, TF10 glass. These fibers integrate a high nonlinearity with a dispersion profile ideally suited for the spectral transformation of Cr: forsterite laser pulses. With the PCF structure designed to provide a nonlinearity on the order of $10^3 \text{ W}^{-1} \text{ km}^{-1}$ at the radiation wavelength of 1 μm and a fundamental-mode dispersion profile with zero group-velocity dispersion around 1.19 μm , unamplified femtosecond Cr: forsterite laser pulses are efficiently frequency-upconverted into the 540–1000-nm wavelength range through solitonic spectral-transformation mechanisms and four-wave mixing. With an array of submicron holes in the fiber core, we tailor the dispersion of PCF modes, providing phase-matched emission of dispersive waves in the desired wavelength region through Cherenkov radiation by solitons.

2. Experimental technique and photonic-crystal fibers

Photonic-crystal fibers used in our experiments were fabricated of TF10 glass using the standard PCF fabrication technology [1,2]. The nonlinear refractive index, n_2 , of TF10 glass is typically an order of magnitude higher than n_2 of fused silica, allowing the enhancement of optical nonlinearity relative to fused silica PCFs with a similar geometry. Material dispersion of TF10 glass also considerably differs from the dispersion of fused silica. In particular, the wavelength λ_z where the group-velocity dispersion (GVD) is zero in the bulk of TF10 glass, $\lambda_z \approx 2.0 \mu\text{m}$, is noticeably shifted toward longer wavelengths relative to the zero-GVD wavelength of fused silica.

The core-cladding geometry of our fiber (Fig. 1) is typical of an index-guiding PCF [1]. The pitch of the structure

Fig. 1. An SEM image of the photonic-crystal fiber: (a) general and (b) close-up view. The scale bar corresponds to 5 μm (a) and 2 μm (b).

in the cladding is 1.4 μm , and the diameter of air holes in the fiber cladding is 1.2 μm . However, in addition to the standard structure of PCF, the core of our PCF features a hexagonal array of six additional air holes with a diameter of about 0.5 μm , running along the fiber axis. These holes serve to provide the desired dispersion properties of waveguide modes [33,34], allowing phase-matched emission of dispersive waves around 680 nm through Cherenkov radiation by solitons, as explained in Section 3. To analyze the properties of modes supported by such a waveguide structure, we employed a modification of the fully vectorial localized-function procedure [35], solving the vectorial wave equations for the electric field $\vec{E}(z, t) = \vec{E} \exp(i(\beta z - ckt))$, where $\vec{E} = (E_x, E_y, E_z)$, β is the propagation constant, k is the wave number, and c is the speed of light in vacuum. The two-dimensional profile of the refractive index $n^2(x, y)$ is approximated with a series expansion in Hermite–Gaussian polynomials and trigonometric functions. The E_x and E_y components of the electric field are represented as series expansions in Hermite–Gaussian polynomials. A substitution of these series expansions for E_x , E_y , and $n^2(x, y)$ into the wave equations for transverse field components reduces the problem to a matrix-equation eigenfunction and eigenvalue problem, allowing the propagation constants and transverse field profiles to be determined for the PCF modes.

In Fig. 2a, we present the field intensity profiles and the wavelength dependence of GVD calculated with the use of this technique for the fundamental and several low-order modes of the fiber. As can be seen from these calculations, the light field in the fundamental and low-order modes of the PCF is confined within the circle bounded by the hexagonal array of air holes. Along with the high nonlinearity of TF10 glass, this strong field confinement provides a high nonlinearity coefficient, $\gamma = (2\pi/\lambda)(\int n_2(x, y) S_z^2 dx dy b / |\int S_z dx dy|^2)$, 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). For the considered PCF, the nonlinear coefficient is estimated as $\gamma \approx 1300 \text{ W}^{-1} \text{ km}^{-1}$ at the wavelength of 800 nm, decreasing to approximately $700 \text{ W}^{-1} \text{ km}^{-1}$ at 1.25 μm .

Fig. 2. Field intensity profiles (a) and group-velocity dispersion (b) for the fundamental (1 and 2) and higher order (3–6) PCF modes.

The implemented PCF design provides an anomalous dispersion for Cr: forsterite laser radiation, with the zero-GVD wavelength λ_z ($\lambda_z \approx 1.19 \mu\text{m}$ for the fundamental mode of our PCF, Fig. 2b) lying close to the central wavelength of Cr: forsterite laser pulses ($1.25 \mu\text{m}$). A fiber with such dispersion properties offers ideal conditions for efficient spectral transformation and frequency shifting of Cr: forsterite laser pulses. The magnitude of optical loss for the fundamental PCF mode was estimated as 12 dB/m at the wavelength of $1.25 \mu\text{m}$.

The laser system used in our experiments was based on a two-arm long-cavity Cr^{4+} : forsterite master oscillator con-

sisting of a 10-mm Cr^{4+} : forsterite crystal, a set of totally reflecting mirrors, an 8% output coupler, and a prism pair for dispersion compensation. The laser oscillator was pumped with a 7.5-W ytterbium fiber laser radiation. The laser cavity was adjusted to generate 70-fs pulses with an average power up to 150 mW and a central wavelength of $1.25 \mu\text{m}$ at a repetition rate of about 20 MHz. The laser pulses were coupled into the PCF through a micro-objective, providing a coupling efficiency of 12%. This coupling efficiency includes losses introduced by an optical isolator, used to reduce the influence of laser radiation retroreflected from the input end of the PCF on the operation of the laser cavity. The mode content of laser radiation in the fiber was modified by changing the tilt angle of the input laser beam with respect to the fiber axis.

3. Results and discussion

Since the central wavelength of Cr: forsterite laser pulses falls within the range of anomalous dispersion (Fig. 2b), these pulses can form solitons in the fundamental and higher order modes of the PCF. High-order dispersion induces wave-matching resonances between solitons and dispersive waves [36,37], giving rise to intense blue-shifted emission, observed as prominent features in the PCF output spectra (Fig. 3a and b). The central wavelength of this blue-shifted emission is controlled by phase matching between the parent soliton and the emitted dispersive wave.

In Fig. 4, we plot the mismatch $\delta\beta = \beta_s(\lambda_0) - \beta(\lambda_d)$ of the propagation constant $\beta(\lambda_d)$ of dispersive-wave emission at the wavelength λ_d in one of the PCF modes and the propagation constant of the soliton $\beta_s(\lambda_0)$ centered at the pump wavelength $\lambda_0 = 1.25 \mu\text{m}$. As can be seen from the comparison of Figs. 3a and 4, the central wavelengths of blue-shifted emission bands observed at the output of the PCF correlate well with the wavelengths where dispersive waves guided in the fundamental (intensity profiles and GVD curve 1 and 2 in Fig. 2a and b) and higher order (intensity profiles and GVD curves 3–6 in Fig. 2a and b) PCF modes are phase-matched with the soliton produced by Cr: forsterite laser pulses. In particular, the $1.25\text{-}\mu\text{m}$ soliton in the fundamental PCF mode is phase-matched with a dispersive wave at 888 nm (curve 1 in Fig. 4). In experimental spectra, this corresponds to the blue-shifted emission band centered around 880–890 nm (curve 1 in Fig. 3a). For the higher order PCF mode labeled as mode 4 in Fig. 2, the soliton-dispersive wave phase matching is achieved at 768 nm, where another intense emission band is observed in the PCF output (curve 2 in Fig. 3a). Finally, the spectral peak centered at 680 nm in the experimental spectra in Fig. 3a is attributed to a wave-matching resonance between the soliton at $1.25 \mu\text{m}$ and mode 6 of the PCF (see Figs. 2 and 4). In experiments, we used a set of color-glass filters to visualize the spatial field intensity profiles corresponding to the dominant spectral peaks in the PCF output. These measurements have confirmed the assignment of modes to the spectral peaks suggested above.

Fig. 3. (a) The blue-shifted output of a 20-cm section of PCF. The laser beam is (1) aligned with the fiber axis and (2) tilted with respect to the fiber axis. (b) The spectrum of radiation transmitted through the TF10 glass PCF with a length of 20 cm. The input pulse energy is 2 nJ. The initial pulse width is 70 fs.

Fig. 4. The propagation-constant mismatch $\delta\beta$ between the soliton centered at 1.25 μm and dispersive waves guided by the fundamental mode of the PCF (1) and modes with intensity profiles 4 (2) and 6 (3), shown in Fig. 2, as a function of the wavelength of the dispersive wave.

With this work largely motivated by the idea of creating compact PCF sources for nonlinear Raman spectroscopy [17,18,20], of special interest is the dispersive-wave emission with a central wavelength at 680 nm (Fig. 4). Along with the femtosecond second-harmonic output of the Cr: forsterite laser, the blue-shifted PCF output centered at 680-nm, employed as a Stokes field in the spectroscopy of coherent anti-Stokes Raman scattering, provides an access to the 1400–1500- cm^{-1} fingerprint region of Raman transitions in molecules of biological significance. Phase matching between the solitons in the infrared part of the spectrum and dispersive waves around 680 nm has been achieved in our experiments by using an array of submicron air holes in the PCF core. The role of submicron air holes in the PCFs employed in our experiments is illustrated in Fig. 5. Dashed line 1 in this figure shows the GVD profile for the waveguide mode 6 (Fig. 2a) of the PCF with nanoholes in the fiber core (Fig. 1a and b and inset 1 in Fig. 5). The counterpart of this mode in a PCF with the same structure, but without nanoholes (inset 2 in Fig. 5) features a dispersion profile shown by dashed line 2 in Fig. 5. To provide phase matching between a soliton and a dispersive wave at a desired wavelength, not only the zero-GVD wavelength, but the whole dispersion profile needs to be optimized, as the propagation-constant mismatch $\delta\beta$ is controlled by high-order dispersion of the guided modes. In the case considered here, while a PCF with properly arranged nanoholes provides phase matching $\delta\beta = 0$ at 680 nm (curve 3 in Fig. 5), an analogous PCF without nanoholes does not allow any soliton-dispersive wave phase matching whatsoever (curve 4 in Fig. 5).

Fig. 5. Dispersion profiles (dashed curves 1 and 2) and the propagation-constant mismatch $\delta\beta$ between a 1.25- μm soliton and a dispersive wave in the PCF mode with intensity profile 6 (3 and 4) for a PCF with (1 and 3) and without (2 and 4) nanoholes modifying the fiber core. PCF structures with and without nanoholes are shown in insets 1 and 2, respectively.

Interestingly and quite importantly, the energy of laser pulses required for the efficient generation of the blue-shifted emission band centered at 880–890 nm is much lower than the energy necessary for the generation of the emission bands centered at 680 and 770–780 nm. This tendency agrees well with the dispersion profiles of these modes. The emission band centered at 880–890 nm is identified as a dispersive-wave emission of the soliton in the fundamental mode. The GVD provided by our PCF for this mode is much lower than the GVD for the higher order modes (see Fig. 2b). This implies the lower radiation energy required for the formation of the fundamental soliton. Indeed, emission bands in higher order modes show up as the input energy increases or the fiber is tilted with respect to the input beam, providing the higher energy input for higher order modes.

The shortest wavelength emission band in the PCF output is centered at 540 nm. This band is observed only in the presence of spectral peaks at 680 and 880–890 nm (Fig. 3). Radiation with frequencies ω_a corresponding to the 540-nm emission band can be generated through four-wave mixing (FWM) $\omega_1 + \omega_2 = \omega_a + \omega_s$, involving Stokes photons (ω_s) from the 880–890-nm emission band and a pair of pump photons (ω_1 and ω_2) from the 680-nm band. A complex beam-pattern structure of 540-nm emission in the PCF output is indicative of multimode phase matching [38] as a mechanism leading to efficient FWM.

In the experiments presented above, frequency conversion of laser radiation predominantly occurs through soliton mechanisms of pulse transformation. The figure of merit provided by a non-silica highly nonlinear PCFs in this regime can be sensibly introduced through the basic soliton parameters. Solitonic phenomena can be conveniently scaled by using the soliton order [38] $N = (\gamma P_0 \tau_0^2 / |\beta_2|)^{1/2}$, where P_0 is the peak power, τ_0 is the soliton pulse width, and $\beta_2 = \partial^2 \beta / \partial \omega^2$ is the parameter characterizing group-velocity dispersion. The power required to generate a fundamental soliton ($N = 1$) is given by $P_0 = |\beta_2| / (\gamma \tau_0^2)$. With the GVD and the pulse width remaining the same, higher- γ fibers would thus require less peak power for the formation of the fundamental soliton. For TF10 glass PCFs considered in this work, the nonlinear coefficient was estimated as approximately $700 \text{ W}^{-1} \text{ km}^{-1}$ at $1.25 \mu\text{m}$ (see Section 2). This value has to be compared with a typical γ of highly nonlinear silica PCFs (typically $70\text{--}90 \text{ W}^{-1} \text{ km}^{-1}$ [39] for the same radiation wavelength of $1.25 \mu\text{m}$). With fixed GVD and pulse width, we thus find that TF10 glass PCFs demonstrated in this work can increase the figure of merit for solitonic mechanisms of frequency conversion by a factor of 7–10 relative to highly nonlinear silica PCFs, allowing the creation of highly efficient fiber-optic frequency shifters for ultrashort laser pulses.

4. Conclusion

We have demonstrated in this work the nonlinear-optical performance of novel PCFs made of high- n_2 TF10

glass. The desired dispersion and nonlinearity properties of guided modes in our PCFs have been achieved with a specifically designed array of nanoholes in the fiber core, allowing efficient frequency upconversion of femtosecond pulses of $1.2\text{--}1.3\text{-}\mu\text{m}$ Cr: forsterite laser radiation. With the PCF structure designed to provide a nonlinearity on the order of $10^3 \text{ W}^{-1} \text{ km}^{-1}$ at the radiation wavelength of $1 \mu\text{m}$ and a fundamental-mode dispersion profile with zero GVD around $1.19 \mu\text{m}$, unamplified femtosecond Cr: forsterite laser pulses have been efficiently frequency-converted into the 540–1000-nm wavelength range through solitonic spectral-transformation mechanisms and FWM. Because of a special importance of this wavelength range for a microscopy and imaging of biological objects [40–42], PCFs demonstrated in this work may allow the creation of compact devices and handpieces for the wavelength-selective imaging of biological tissues, including optical coherence tomographs, nonlinear-optical microscopes, and fiber endoscopes.

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