Phase-matched four-wave mixing of sub-100-TW/cm^2 femtosecond laser pulses in isolated air-guided modes of a hollow photonic-crystal fiber

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Hollow-core photonic-crystal fibers are shown to allow propagation and nonlinear-optical frequency conversion of high-intensity ultrashort laser pulses in the regime of isolated guided modes confined in the hollow gas-filled fiber core. With a specially designed dispersion of such modes, the $3\omega=2\omega+2\omega-\omega$ four-wave mixing of fundamental (ω) and second-harmonic (2ω) sub-100-TW/cm^2 femtosecond pulses of a Cr:forsterite laser can be phase matched in a hollow photonic-crystal fiber within a spectral band of more than 10 nm, resulting in the efficient generation of femtosecond pulses in a well-resolved higher-order air-guided mode of 417-nm radiation.

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I. INTRODUCTION

Nonlinear optics of high-intensity ultrashort laser pulses is one of the most interesting and rapidly growing areas of optical physics. Nonlinear-optical interactions of such pulses lead to the generation of high-order optical harmonics [1,2], result in the filamentation and self-channeling of laser radiation [3], and allow the synthesis of attosecond-field waveforms [4–6]. The standard method of achieving high intensities of laser radiation involves focusing amplified output pulses of high-power laser systems. The effective length of nonlinear-optical interactions is limited in this geometry to the confocal parameter, which is equal to $b=n\omega w_0^2/c$ for a Gaussian beam with a radius w_0 (ω is the radiation frequency and n is the refractive index). Hollow fibers [7] offer attractive guided-wave solutions for strong-field nonlinear optics, allowing the effective interaction length to be substantially increased [8,9]. Since the laser breakdown threshold for gases filling the core of a hollow fiber is typically several orders of magnitude higher than the breakdown threshold for standard, solid-cladding fibers, hollow-core fibers are intensely used for the compression of high-intensity laser pulses through Kerr-nonlinearity-induced self-phase-modulation [10,11] and high-order stimulated Raman scattering [12], frequency conversion through wave mixing [9,13] and optical harmonic generation [14–16], and high-sensitivity four-wave mixing spectroscopy [8,17].

The air-guided modes in standard hollow fibers are leaky, with the magnitude of losses scaling as [7] λ^2/a^3 with the fiber inner radius a and the radiation wavelength λ, which dictates the choice of hollow fibers with $a\sim 50$–300 μm for nonlinear-optical experiments. Such large-a fibers are essentially multimode, with many guided modes typically contributing to nonlinear-optical interactions [9,17]. The number of air-guided modes involved in the transportation of laser radiation is radically reduced in the case of hollow-core photonic crystal fibers (PCFs) [18,19]. Such fibers guide light due to the high reflectivity of a two-dimensionally periodic (photonic-crystal) cladding (the inset in Fig. 1) within photonic band gaps (PBGs). Low-loss guiding in a few or even a single air-guided mode can be implemented under these conditions in a hollow core with a typical diameter of 10–20 μm [18–20]. Hollow PCFs with such core diameters have been recently demonstrated to enhance nonlinear-optical processes, including stimulated Raman scattering [21], four-wave mixing (FWM) [22], and self-phase-modulation [23]. Air-guided modes in hollow PCFs can support megawatt optical solitons [24] and allow transportation of high-intensity laser pulses for technological [25] and biomedical [26] applications.

In this work, we show that hollow-core PCFs provide a unique opportunity of guiding and frequency converting of high-intensity ultrashort laser pulses in the regime of isolated, well-defined air-guided modes. While standard, solid-core fibers get damaged by high-intensity laser radiation above the self-focusing and optical breakdown thresholds and conventional solid-cladding hollow waveguides are essentially multimode because of the reasons outlined above,

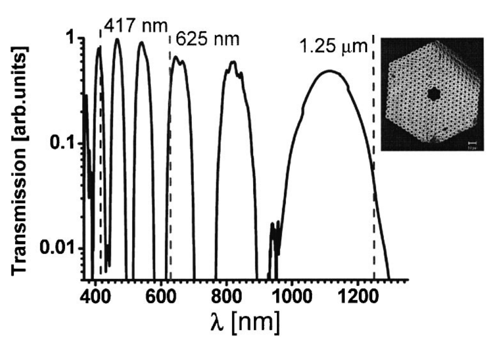

FIG. 1. Transmission spectrum of the hollow PCF designed to simultaneously support air-guided modes of the 1.25-μm fundamental radiation of the Cr:forsterite laser, as well as its second (625 nm) and third (417 nm) harmonics. The wavelengths of the fields involved in the four-wave mixing process are shown by dashed lines. The inset shows an image of the PCF cross section.

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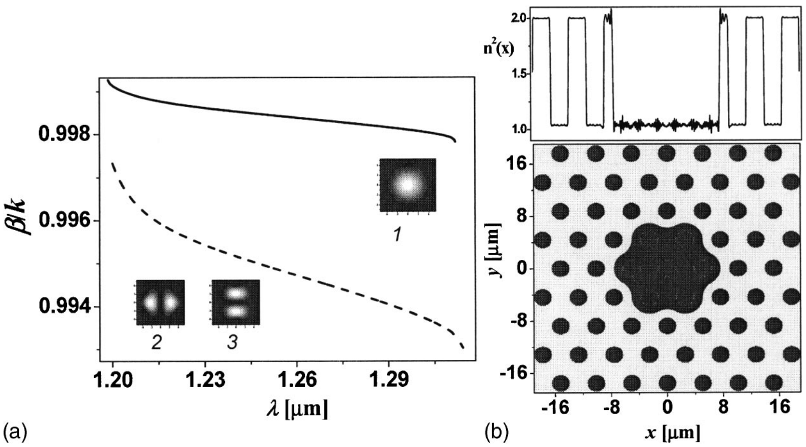

FIG. 2. (a) Propagation constant β normalized to the wave number k as a function of the wavelength for the fundamental (solid curve) and a doublet of higher order (dashed curve) air-guided modes of a hollow PCF with a core diameter of approximately 13 μm and a period of the photonic-crystal cladding of about 4.6 μm. The insets show intensity profiles for the fundamental (1) and the doublet of higher-order air-guided modes (2 and 3) of the hollow PCF. (b) The profile of $n^2(x,y)$ in the cross section of a hollow PCF synthesized with 80×80 Hermite-Gaussian polynomials and 150×150 trigonometric functions: (top) 1D cut; (bottom) 2D profile shown by levels of gray scale.

hollow PCFs can support isolated, robust, well-defined truly waveguide modes of electromagnetic radiation in a gas-filled hollow core only a few micrometers in diameter [18–21]. We will demonstrate in what follows that these isolated air-guided modes of high-intensity ultrashort pulses can be efficiently frequency converted in hollow PCFs through enhanced waveguide FWM, withstanding laser energy densities well above the self-focusing and laser breakdown thresholds for all the available nonlinear-optical crystals and conventional fibers. In earlier work [22], hollow PCFs have been shown to allow a substantial waveguide enhancement of FWM for narrowband, picosecond laser pulses with relatively low intensities. Our goal here is to extend this approach to a qualitatively different regime by phase-matching the FWM of isolated PCF modes within a broad spectral range for broadband, femtosecond pulses with intensities on the order of 100 $\mathrm{TW/cm^2}$, corresponding to laser energy densities exceeding typical breakdown thresholds for standard nonlinear crystals and conventional fibers. A specially designed dispersion of modes in hollow PCFs will be shown to allow a phase matching to be achieved for the $3\omega=2\omega+2\omega-\omega$ four-wave mixing of fundamental (ω) and second-harmonic (2ω) sub-100-$\mathrm{TW/cm^2}$ femtosecond pulses of a Cr:forsterite laser, resulting in the efficient generation of a well-resolved higher-order air-guided mode of 420-nm radiation.

II. GAS-FILLED HOLLOW PHOTONIC-CRYSTAL FIBERS: DISPERSION AND PHASE MATCHING

Hollow-core PCFs designed for the purposes of these experiments had an inner diameter of approximately 13 μm and a period of the photonic-crystal cladding of about 4.6 μm. A typical structure of the PCF cross section is shown in the inset to Fig. 1. The PCFs were fabricated [20,25] with the use of a preform consisting of a set of identical glass capillaries. Seven capillaries were removed from the central part of the preform for the hollow core of PCFs. Transmission spectra of these hollow-core PCFs displayed characteristic well-pronounced isolated passbands (Fig. 1), related to PBGs of the cladding [18–20,27]. Our PCFs were designed in such a way that all the three field components (with central frequencies ω, 2ω, and 3ω) fall inside the fiber passbands (Fig. 1), related to the photonic band gaps of the photonic-crystal cladding. Guiding by photonic band gaps [18,19] is thus the mechanism of waveguiding for all the fields involved in the FWM process under study. The magnitude of optical losses at 1.25 μm for these PCFs was estimated as 0.09 $\mathrm{cm^{-1}}$.

The dispersion of air-guided modes in hollow PCFs [Fig. 2(a)] was tailored by changing the fiber cladding structure [25]. To design the dispersion of PCF modes allowing phase-matched FWM of the fundamental and second-harmonic femtosecond pulses of a Cr:forsterite laser, we developed a numerical procedure solving the vectorial wave problem 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, and k is the wave number:

$$\left[\frac{\nabla_\perp^2}{k^2}+n^2(x,y)\right]E_{x,y}+\frac{1}{k^2}\left(E_x\frac{\partial\ln(n^2)}{\partial x}+E_y\frac{\partial\ln(n^2)}{\partial y}\right)'_{x,y}=\frac{\beta^2}{k^2}E_{x,y}. \quad (1)$$

Here, $\nabla_\perp$ is the gradient in the (x,y) plane, $n=n(x,y)$ is the two-dimensional profile of the refractive index, and the prime in the second term on the left-hand side stands for differentiation. The profile of $n^2(x,y)$ was approximated, as in [28], with a series expansion in Hermite-Gaussian polynomials and trigonometric functions. Figure 2(b) displays a one-dimensional (1D) cut (top) and a 2D profile (bottom) of $n^2(x,y)$ synthesized with 80×80 Hermite-Gaussian polynomials and 150×150 trigonometric functions. The E_x and E_y components of the electric field were represented as series expansions in Hermite-Gaussian polynomials. A substitution of the series expansions for E_x, E_y, and $n^2(x,y)$ into Eq. (1) reduces the problem to an eigenfunction and eigenvalue problem of a matrix equation, which allows the propagation constants and transverse field profiles to be determined for the air-guided modes of hollow PCFs.

Figure 2(a) illustrates dispersion properties and presents typical field intensity profiles for the fundamental and higher-order guided modes in our hollow PCFs. The fundamental mode has the maximum propagation constant β [solid

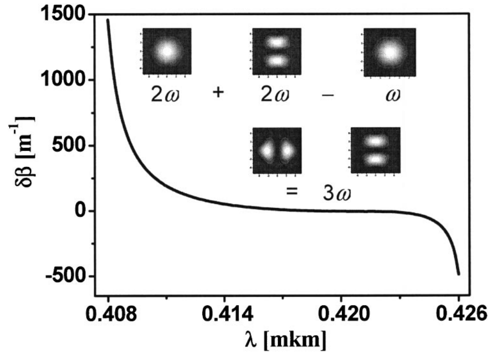

FIG. 3. The group-velocity mismatch calculated for the four-wave mixing $3\omega=2\omega+2\omega-\omega$ of the fundamental mode of 1.25-μm Cr:forsterite laser radiation (ω), the fundamental and one of the higher-order modes of the second-harmonic pump field (2ω), and a higher-order mode of the FWM signal (3ω) in a hollow PCF with an inner diameter of approximately 13 μm and a period of the photonic-crystal cladding of about 4.6 μm. Field intensity distributions in the air-guided modes of the hollow PCF involved in the considered FWM process are shown in the inset.

curve in Fig. 2(a)], and the electric-field intensity in this mode reaches its maximum at the center of the fiber core, monotonically decreasing off the center of the fiber [inset 1 in Fig. 2(a)]. Higher-order modes form degenerate multiplets [insets 2 and 3 in Fig. 2(a)], with their superposition supporting the full symmetry of the fiber. Dispersion of a doublet of higher-order air-guided modes of a hollow PCF is shown by the dashed line in Fig. 2(a). Waveguide modes of this type will be used in our experiments to phase-match the FWM process in a hollow PCF.

We now consider an FWM process $3\omega=2\omega+2\omega-\omega$ involving the fundamental mode of 1.25-μm Cr:forsterite laser radiation (ω), the fundamental and one of the higher-order modes of the second-harmonic pump field (2ω), and a higher-order mode of the FWM signal (3ω). This combination of waveguide modes with different central frequencies is illustrated in the insets to Fig. 3. We mix the fundamental waveguide mode of the first second-harmonic pump field involved in the FWM process (labeled by 2ω in the inset to Fig. 3) with a higher order waveguide mode with the same carrier frequency (also shown in the inset to Fig. 3) and the fundamental PCF mode of fundamental radiation of the Cr:forsterite laser (the field ω in the inset to Fig. 3). The phase-matched four-wave mixing of these fields gives rise to a doublet of higher-order PCF modes with the carrier frequency 3ω (field intensity profiles for this doublet of modes are shown in the lower line in the inset to Fig. 3). In a hollow PCF with an ideal sixth order symmetry, the guided modes of this doublet are degenerate. In the analysis presented in this work, we neglect deviations of the PCF structure from this ideal geometry, which in practice remove the degeneracy of modes.

Figure 3 presents the group-velocity mismatch $\delta\beta=\beta_{3\omega}-\beta'_{2\omega}-\beta''_{2\omega}+\beta_{\omega}$ calculated for such an FWM process in air-guided modes of the hollow PCF (β_{ω}, $\beta'_{2\omega}$ $\beta''_{2\omega}$, and $\beta_{3\omega}$ are the propagation constants of the air-guided modes of the fundamental, second-harmonic, and FWM signal fields in the hollow PCF). As can be seen from Fig. 3, a nearly perfect phase matching can be achieved for the FWM of the above-specified air-guided modes of the hollow PCF within a spectral band with a width exceeding 10 nm. Such a PCF design is, therefore, ideally suited for an efficient nonlinear-optical frequency conversion of femtosecond pulses of a Cr:forsterite laser.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The laser system employed in our experiments (Fig. 4) 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 fiber ytterbium laser, generated (30–50)-fs light pulses with 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-fs pulse duration with the maximum laser pulse energy up to 30 μJ at 1 kHz. A 1-mm-thick BBO crystal was used to generate the second harmonic of amplified Cr:forsterite laser radiation.

Femtosecond pulses of 1.25-μm fundamental radiation and 625-nm second-harmonic radiation with pulse energies ranging from 0.1 up to 10 μJ were used as pump fields ω and 2ω in the FWM process. These pulses were coupled into the hollow PCF, placed on a three-dimensional translation stage, with a standard micro-objective. Fundamental radiation was guided in the fundamental mode of the PCF (inset 1 in Fig. 5), while the second harmonic was coupled into a mixture of the fundamental and higher-order air-guided modes (inset 2 in Fig. 5). The FWM of these two pump fields induced by the third-order nonlinearity of the atmospheric-pressure air filling the PCF resulted in the generation of a signal centered at the frequency of the third harmonic, 3ω, of fundamental radiation of the Cr:forsterite laser. The signal at this frequency

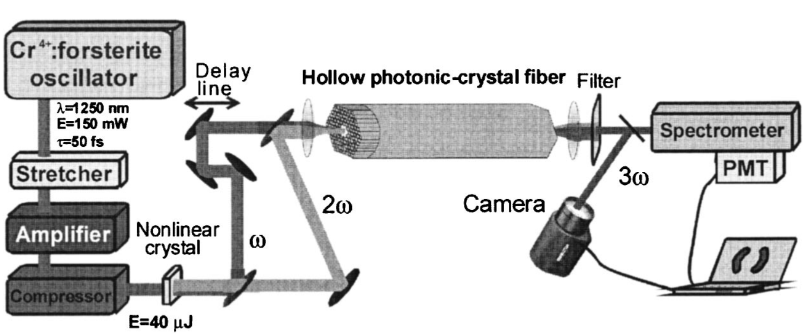

FIG. 4. Diagram of four-wave mixing of high-intensity femtosecond laser pulses in a hollow photonic-crystal fiber.

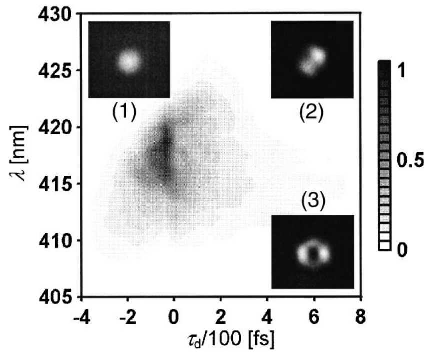

FIG. 5. The spectra of the FWM signal measured at the output of a 6-cm hollow PCF for different delay times τ_d between the input pump pulses. The energies of the fundamental and second-harmonic pulses at the input of the PCF are 2 and 4 μJ, respectively. The initial duration of the pump pulse of fundamental radiation is 50 fs. The insets show the spatial beam profiles measured for the fundamental (1) and second-harmonic (2) pump beams and the FWM signal (3) at the output of the PCF.

can be produced in a hollow-core PCF through both the $3\omega=2\omega+2\omega-\omega$ FWM process and direct third-harmonic generation $3\omega=\omega+\omega+\omega$. Experiments performed with only the fundamental beam used as a pump have shown, however, that direct third-harmonic generation is much less efficient than two-color FWM.

The FWM signal with a central wavelength of 417 nm is generated in the hollow PCF in a stable isolated well-resolved higher-order air-guided mode (inset 3 in Fig. 5). Superposition of the doublet of air-guided modes of 3ω radiation, which is nearly perfectly phase-matched with the PCF modes of the two-color pump field (the inset in Fig. 3), provides an ideal fit for the spatial beam profile of the FWM signal observed at the output of the PCF. This result suggests that the spatial beam profile of the FWM signal generated in a PCF as a result of nonlinear-optical interaction of isolated air-guided modes of pump radiation is dictated and stabilized by phase-matching conditions. This remarkable property of FWM in a hollow PCF provides a high beam quality of the nonlinear signal at the output of the fiber, eliminates mode cross-talk, and offers much promise for nonlinear-optical processing of high-intensity laser pulses. The maximum efficiency of FWM frequency conversion achieved in our experiments is estimated as 0.2% for a 6-cm hollow PCF. This FWM efficiency is certainly much lower than typical efficiencies of frequency conversion that can be achieved with nonlinear crystals. However, self-focusing and optical breakdown limit the use of nonlinear crystals for the frequency conversion of high-intensity ultrashort laser pulses. We believe that phase-matched nonlinear-optical interactions in hollow-core PCFs can offer interesting alternatives to standard strategies of frequency conversion using nonlinear crystals at least for high-intensity laser radiation. Larger interaction lengths and therefore higher efficiencies of frequency conversion can be achieved for nonlinear-optical processes in hollow PCFs by optimizing dispersion profiles of guided modes and using mixtures of gases with higher optical nonlinearities and appropriate dispersion-compensating [29] additives.

The spatial walk-off of light pulses involved in the FWM process is one of the main physical factors limiting the efficiency of frequency conversion in the PCF under our experimental conditions. Figure 5, showing the spectra of the FWM signal measured for different delay times τ_d between the input pump pulses, demonstrates a high sensitivity of the efficiency of the FWM process to the group-delay-induced spatial separation of the pump pulses. Group-velocity matching would thus allow a further improvement of the efficiency of nonlinear-optical frequency conversion of femtosecond pulses in hollow PCFs. The spatial beam profile of the FWM signal at the output of the PCF remained stable up to the energy of input pump pulses of about 6 μJ, corresponding to the light-field intensity of about 90 $\mathrm{TW/cm^2}$. Spatial self-action and ionization effects started to play a noticeable role above this level of input laser intensities, leading to instabilities and distortions in output beam profiles of the pump and FWM fields. Laser pulses with energies exceeding 10 μJ produced an optical damage on PCF inner walls in our experiments, resulting in an irreversible degradation of fiber transmission.

IV. CONCLUSION

We have demonstrated in this work that hollow-core photonic-crystal fibers allow propagation and nonlinear-optical frequency conversion of high-intensity ultrashort laser pulses in the regime of isolated guided modes confined in the hollow gas-filled fiber core. These fibers provide a unique opportunity of efficient gas-phase frequency conversion for high-intensity ultrashort laser pulses with energy densities exceeding the optical breakdown threshold for the available nonlinear crystals and conventional fibers. With a specially designed dispersion of such modes, the $3\omega=2\omega+2\omega-\omega$ four-wave mixing of fundamental and second-harmonic sub-100-$\mathrm{TW/cm^2}$ femtosecond pulses of a Cr:forsterite laser can be phase-matched in a hollow photonic-crystal fiber within a spectral band of more than 10 nm, resulting in the efficient generation of femtosecond pulses in a well-resolved higher-order air-guided mode of 420-nm radiation. The ability of hollow PCFs to efficiently phase-match nonlinear-optical interactions of high-intensity ultrashort laser pulses and controlled dispersion of these fibers offer much promise for the enhancement of high-order nonlinear-optical processes, including high-order harmonic generation and high-order stimulated Raman scattering, suggesting attractive solutions for the synthesis of ultrashort field waveforms and efficient generation of short-wavelength radiation.

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