Phase-matched waveguide four-wave mixing scaled to higher peak powers with large-core-area hollow photonic-crystal fibers

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Hollow photonic-crystal fibers with large core diameters are shown to allow waveguide nonlinear-optical interactions to be scaled to higher pulse peak powers. Phase-matched four-wave mixing is predicted theoretically and demonstrated experimentally for millijoule nanosecond pulses propagating in a hollow photonic-crystal fiber with a core diameter of about 50 μm, suggesting the way to substantially enhance the efficiency of nonlinear-optical spectral transformations and wave mixing of high-power laser pulses in the gas phase.

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Progress in high-field physics and advances in high-power laser technologies call for new efficient waveguide solutions. Hollow waveguides [1] have been shown to allow guiding of high-power laser radiation [2] and demonstrated an outstanding performance in the generation of few-cycle laser pulses [3,4], as well as enhanced four-wave mixing [5,6] and high-order harmonic generation [7,8]. However, these fibers can provide tolerable levels of losses only in the multimode regime, allowing no scalability to smaller core diameters, since guiding losses of such fibers increase as a^{-3} with a decrease in the core diameter a [1].

Filamentation of high-intensity ultrashort laser pulses [9] is another interesting waveguiding option, allowing the channeling of terawatt-level laser radiation over several kilometers and offering unique possibilities in remote sensing [10]. Laser beam dynamics in filaments is, however, very complicated. The light field generally undergoes uncontrollable amplitude- and phase-profile transformations and distortions in this regime, leaving apparently no way for a selective manipulation of individual guided modes, as many applications would require.

Hollow-core photonic crystal fibers (PCFs) [11–13] resolve this conflict between the magnitude of radiation losses and the number of air-guided modes. PCFs 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 [12–15]. Hollow PCFs with such core diameters have been recently demonstrated to enhance nonlinear-optical processes, including stimulated Raman scattering [16], four-wave mixing (FWM) [17], coherent anti-Stokes Raman scattering (CARS) [18], and self-phase modulation [19]. The spatial self-action of intense ultrashort laser pulses gives rise to interesting waveguiding regimes in hollow PCFs below the blowup threshold [20]. The SPM-induced spectral broadening of femtosecond laser pulses in air-guided modes of hollow PCFs allows the creation of fiber-optic diodes [21] and limiters [22] for high-intensity ultrashort laser pulses. Air-guided modes in hollow PCFs can support megawatt optical solitons [23] and allow femtosecond soliton pulse delivery over several meters [24], as well as transportation of high-energy laser pulses for technological [25,26] and biomedical [27] applications.

Several designs of hollow PCFs have been demonstrated, allowing the fiber structure and, hence, fiber dispersion and mode profiles to be adapted to a particular application. The dominating design solutions include PCFs with a two-dimensionally periodic hexagonal (honeycomb) photonic-crystal lattice, first demonstrated by Cregan *et al.* [11], and Kagomé-lattice-cladding PCFs, introduced by Benabid *et al.* [16]. The typical size of the PCF core in most of the nonlinear-optical experiments published so far ranges from 6 up to 20 μm. It would be interesting and important for many applications to extend the PCF architecture to hollow fibers with larger inner diameters. Such hollow PCFs would allow higher peak powers to be coupled into guided modes without irreversible damage on fiber walls. As the maximum peak power leading to no damage of PCF scales as $\sim a^2$ with the fiber core diameter a for given pulse duration and threshold

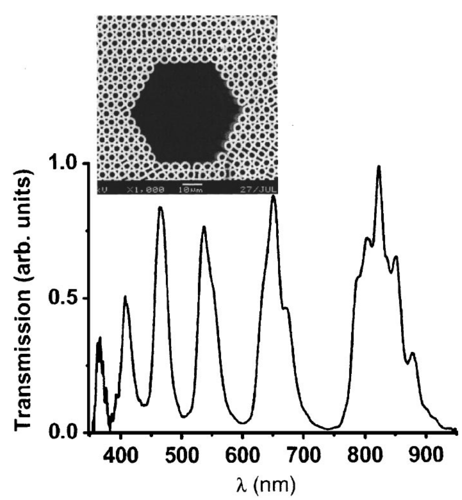

FIG. 1. Transmission spectrum of the hollow-core PCF. The inset shows the cross-section view of the PCF with a period of the cladding structure of about 5 μm.

fluence of optical breakdown, large-core hollow PCFs would suggest a valuable option for the peak-power scaling of pulse-compression, frequency-conversion, and soliton-transmission regimes in the high-intensity ultrafast optics of guided waves, opening the ways for enhanced high-order nonlinear optics, including high-order harmonic generation.

In this work, we show that hollow PCFs with large core diameters can bridge the gap between standard, solid-cladding hollow fibers and previously reported hollow PCFs in terms of effective guided-mode areas, offering attractive solutions for high-field physics, laser technologies, and ultrafast photonics.

PCFs used in our experiments had a hollow core in the form of a regular hexagon with each side corresponding to five cane diameters (the fabrication procedure is described in greater detail elsewhere [28]). The inset in Fig. 1 shows an image of a hollow PCF with a period of the cladding of approximately 5 μm and a core diameter of about 50 μm. Transmission spectra of our large-core PCFs display well-pronounced passbands (Fig. 1), indicating the PBG guidance of radiation in air modes of the fiber.

To model guided modes and transmission spectra of hollow PCFs, we numerically solved the wave equations for the transverse components of the electric field using a modification of the technique developed by Poladian *et al.* [29] (see Ref. [30] for the details of our numerical procedure). Material dispersion was included through the dispersion of atmospheric pressure air in the fiber core and by using the reference data [31] for the wavelength dependence of the refractive index of S93-1 glass, employed as PCF cladding material (see also Ref. [32]). Geometric parameters of the PCF structure were defined from SEM images (inset in Fig. 1), yielding the inner diameter of holes in the cladding of 2.6 μm, the period of the cladding of 5.1 μm, and the distance between the opposite sides of the hexagonal hollow core of the PCF equal to approximately 50 μm. With material dispersion and geometric parameters of PCF structure included in the model as described above, our simulation procedure did not involve any parameter fitting. Because of structure imperfections, the spatial profile of the refractive index in the real PCF (the inset in Fig. 1) slightly deviates from the synthesized model refractive-index profile. Typical calculation errors for the FWM phase-matching wavelength resulting from this deviation, however, did not exceed 1.5 nm.

In the inset to Fig. 2, we plot the propagation-constant mismatch $\delta\beta = \beta_a - 2\beta_1 + \beta_2$ calculated as a function of the tunable-laser wavelength λ_1 for the $\omega_a = 2\omega_1 - \omega_2$ FWM processes in the fundamental mode of the hollow PCF, which has a characteristic bell-shaped spatial intensity profile. The wavelength $\lambda_1 = 2\pi c/\omega_1$ provided by a dye laser (tunable in our experiments from 630 to 665 nm) is mixed in this process with the fixed-frequency field of fundamental Nd:YAG-laser radiation at $\lambda_2 = 2\pi c/\omega_2 = 1064$ nm, to generate an FWM signal within the range of wavelengths $\lambda_a = 2\pi c/\omega_a$ from 445 to 485 nm (β_1, β_2, and β_a stand for the propagation constants at the frequencies ω_1, ω_2, and ω_a, respectively). Around $\lambda_1 = 650$ nm, the propagation constant mismatch passes through zero, implying phase matching for the considered FWM process and providing optimal condi-

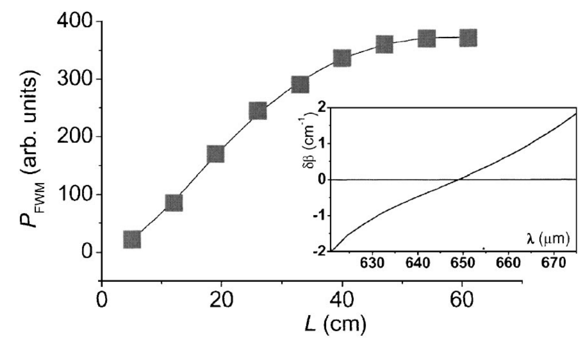

FIG. 2. The power of the four-wave mixing signal P_{FWM} at $\lambda_1 = 650$ nm measured as a function of the PCF length l. The inset shows the mismatch of the propagation constants $\delta\beta = \beta_a - 2\beta_1 + \beta_2$ for air-guided modes in a hollow fiber involved in the $\omega_a = 2\omega_1 - \omega_2$ FWM process as a function of the tunable-laser wavelength λ_1 with $\lambda_2 = 2\pi c/\omega_2 = 1064$ nm. The horizontal line shows the phase-matching condition $\delta\beta = 0$.

tions for efficient FWM frequency conversion. The standard theory of FWM [33,34] predicts the scaling law $P_{FWM} \propto |\chi^{(3)}_{eff}|^2 P_1 P_2^2 M$ for the power P_{FWM} of the FWM signal in this regime, with P_1 and P_2 being the powers of the fields with frequencies ω_1 and ω_2, respectively; $\chi^{(3)}_{eff}$ representing the effective combination of cubic nonlinear-optical susceptibility tensor components; and the factor M including optical losses in the fiber:

$$M = M(\Delta\alpha l, \alpha_a l, \delta\beta l = 0) = \exp[-(\Delta\alpha + \alpha_a)l] \times [\sinh(\Delta\alpha l/2)/(\Delta\alpha l/2)]^2 l^2,$$

$\Delta\alpha = (2\alpha_1 + \alpha_2 - \alpha_a)/2$, α_1, α_2, and α_a are the magnitudes of optical losses at frequencies ω_1, ω_2, and ω_a, respectively, and l is the fiber length. The FWM signal power grows quadratically with the fiber length l as long as l is much less than the optimal fiber length $l_{opt} = (\Delta\alpha)^{-1}\ln[(2\alpha_1 + \alpha_2)/\alpha_a]$, which provides the maximum efficiency of FWM. Waveguide losses should thus be expected to be a dominant mechanism limiting the efficiency of the $\omega_a = 2\omega_1 - \omega_2$ FWM process in the studied PCF for the above-specified set of pump, probe, and signal wavelengths. Results of experiments presented below in this paper confirm the theoretical prediction of phase matching for FWM in a PCF with the above-specified set of pump, probe, and signal wavelengths.

The nanosecond laser system employed in our experiments consisted of a *Q*-switched Nd:YAG master oscillator, Nd:YAG amplifiers, frequency-doubling crystals, a dye laser, as well as a set of totally reflecting and dichroic mirrors and lenses adapted for the purposes of FWM experiments. The *Q*-switched Nd:YAG master oscillator generated 15-ns pulses of 1064-μm radiation, which were then amplified up to about 30 mJ by Nd:YAG amplifiers. A KDP crystal was used for the frequency doubling of the fundamental radiation. This second-harmonic radiation served as a pump for the dye laser, generating frequency-tunable radiation within the wavelength range from 630 to 670 nm. The fundamental-wavelength output and frequency-tunable dye-laser radiation were employed as pump fields in the $\omega_a = 2\omega_1 - \omega_2$ FWM

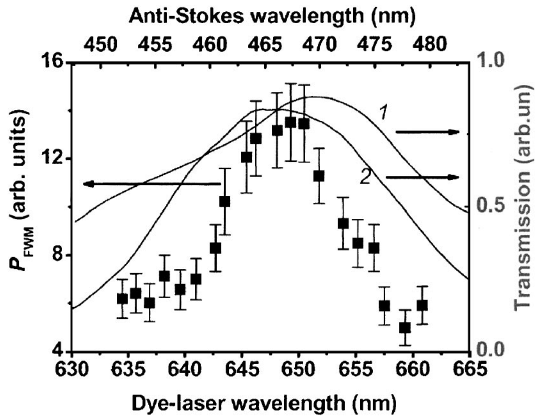

FIG. 3. (Color online). Power of the $\omega_a=2\omega_1-\omega_2$ four-wave mixing signal from the hollow PCF with the length of 30 cm versus the wavelengths of dye-laser (lower axis) and FWM anti-Stokes signal (upper axis) radiation; $\lambda_2=1064$ nm. Fiber transmission for dye-laser radiation (solid line *1*) and the FWM anti-Stokes signal (solid line *2*) is also shown.

process, as described below. The frequency dependence of the FWM signal produced through the nonlinear-optical interaction in the hollow PCF was measured point by point by scanning the frequency of dye-laser radiation. The energies of these pump fields were varied in our experiments from 0.5 up to 10 mJ at the fundamental wavelength and from 0.05 to 0.7 mJ for dye-laser radiation. The PCF could withstand the energy of fundamental radiation up to 10 mJ, corresponding to a laser fluence of approximately 630 J/cm^2, without an irreversible degradation of fiber performance because of optical breakdown.

In the FWM process studied in our experiments, two pump waves with the wavelength $\lambda_1=2\pi c/\omega_1$ ranging from 630 to 665 nm, provided by the dye laser, are mixed with the fixed-frequency field of the fundamental radiation at $\lambda_2=1064$ nm, to generate an anti-Stokes signal within the range of wavelengths λ_a from 445 to 485 nm. The spectrum of the anti-Stokes signal at the output of the PCF displays a maximum at $\lambda_1\approx 650$ nm (Fig. 3), which exactly corresponds to the wavelength where the phase matching is achieved for the considered type of the FWM process (the inset in Fig. 2).

To check the phase-matched nature of FWM in our PCF experiments, we measured the power of the FWM signal P_{FWM} at $\lambda_1=650$ nm as a function of the PCF length *l*. For this purpose, we used a hollow PCF with a length of 61 cm and gradually decreased the length of the fiber. Waveguide losses at the wavelengths of the pump, probe, and signal fields were estimated as $\alpha_1\approx\alpha_a\approx 0.014\ \text{cm}^{-1}$ and $\alpha_2\approx 0.04\ \text{cm}^{-1}$ for the PCF sample used in these experiments. The experimentally measured dependence of P_{FWM} on *l* (Fig. 2) for constant powers of the pump and probe fields is nearly ideally fitted by the function $M(\Delta\alpha l,\alpha_a l,\delta\beta l=0)$ with $\Delta\alpha=0.025\ \text{cm}^{-1}$ and $\alpha_a=0.01\ \text{cm}^{-1}$. In agreement with theoretical predictions, the FWM power scales as l^2 as long as waveguide losses remain negligible for small *l*. With the PCF length approaching $l_{opt}\approx 58$ cm, P_{FWM} saturates and then falls off. These results show that PCF losses at the frequency of fundamental Nd:YAG laser radiation is the dominant factor limiting the FWM efficiency in our PCF. The maximum measured conversion efficiency for the FWM process, defined as the ratio of P_{FWM} to the power P_1 of dye-laser radiation, was equal to 7×10^{-6} for the energies of dye-laser and fundamental radiation fields equal to 0.7 and 9 mJ, respectively. This result agrees by its order of magnitude with theoretical predictions $P_{FWM}/P_1\approx 10^{-5}$, obtained with a standard value of nonresonant hyperpolarizability of atmospheric air, $5\times 10^{-38}\ \text{cm}^6/\text{erg}$ [33]. The observed behavior of P_{FWM} as a function of *l* allows the lower bound for the coherence length $l_c=\pi/(2|\delta\beta|)$ for the considered FWM process to be quantified: $l_c>l_{opt}\approx 58$ cm. The coherence length for waveguide FWM in our PCF experiments has been thus increased, with an appropriate design of the PCF dispersion profile, by more than an order of magnitude with respect to the coherence length of the same FWM process in the regime of tight focusing in the same gas without a waveguide (a few centimeters for the atmospheric-pressure air and the chosen set of laser field wavelengths).

We have shown in this work that large-core-area hollow PCFs bridge the gap between standard, solid-cladding hollow fibers, and hollow PCFs in terms of effective guided-mode areas, allowing peak-power scaling of phase-matched waveguide four-wave mixing of laser pulses. Improved phase matching has been predicted theoretically and demonstrated experimentally for waveguide FWM in a hollow PCF with a core diameter of about 50 μm, suggesting a way to substantially enhance the efficiency of nonlinear-optical wave mixing of high-power laser pulses in the gas phase. Interesting extensions of PCF-based phase-matching strategies demonstrated in this work to higher order nonlinear-optical processes in the gas phase include high-order harmonic generation and multiwave mixing.

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