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Hollow-core photonic-crystal fibers optimized for four-wave mixing and coherent anti-Stokes Raman scattering

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Hollow-core photonic-crystal fibers optimized for four-wave mixing (FWM) and coherent anti-Stokes Raman scattering (CARS) spectroscopy were designed and fabricated. These fibers provide maximum transmission for a two-color pump and the FWM signal, allowing FWM to be enhanced by a factor of ∼800 compared with the tight-focusing regime. A reliably detectable FWM signal was generated under these conditions with micro- and even submicrojoule picosecond pump pulses, i.e. at the level of pump energies unprecedentedly low for non-resonant FWM in the gas phase. The FWM efficiency is shown to depend on the relative orientation of polarization vectors of input pump pulses, offering the extension of FWM and CARS polarization techniques to air-guided modes in photonic-crystal fibers. Copyright © 2003 John Wiley & Sons, Ltd.

KEYWORDS: four-wave mixing; fibers; photonic crystals

INTRODUCTION

Waveguide optics provides efficient means for enhancing non-linear optical interactions and improving the sensitivity of nonlinear spectroscopy. The efficiency of four-wave mixing (FWM) is improved in the waveguide regime[1–9] owing to a larger interaction length and higher intensities of pump waves achieved for given pump powers by reducing the transverse size of the waveguiding layer in planar waveguides or the core diameter in optical fibers. Waveguide regimes expand the abilities of coherent anti-Stokes Raman scattering (CARS)[2–5,9] and general-type FWM,[6–8] offering the ways to increase substantially the level of the non-linear signal,[2,6] suppress the non-resonant background in CARS[3,5] and even attack the monolayer sensitivity by using CARS spectroscopy of molecules on surfaces and interfaces.[5] Hollow waveguides and fibers, as demonstrated by pioneering experiments on coherent anti-Stokes Raman scattering performed by Miles *et al.*[2] and subsequent work on general-type FWM,[6,8,9] are ideally suited for the FWM spectroscopy of the gas phase. Hollow fibers have also been shown to permit efficient pulse compression through self-phase modulation[10,11] and high-order stimulated Raman scattering,[12] as well as frequency conversion and control of ultrashort pulses through parametric wave mixing and cross-phase modulation.[13,14]

The waveguide FWM enhancement factor scales[2] as $\lambda^2 l^2/a^4$ with radiation wavelength λ, the fiber length l and the fiber inner radius a, allowing the FWM efficiency to be substantially improved with respect to the regime of tightly focused laser beams. The interaction length l is limited in the case of hollow fibers by optical losses, whose magnitude is proportional[15] to λ^2/a^3. The waveguide FWM enhancement factor also becomes limited under these conditions, scaling as $\lambda^2/\alpha^2 a^4$. Optical losses, growing with a decrease in the inner radius a, limit FWM enhancement, with the FWM enhancement factor rapidly declining with decreasing a for small values of the fiber inner radius.

The situation radically changes in the case of recently demonstrated[16] photonic-crystal hollow fibers—hollow-core fibers with a two-dimensionally periodic microstructure

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cladding (two-dimensional photonic crystal). Photonic bandgaps in transmission spectra of a two-dimensional periodic cladding in these fibers provide high reflection coefficients for electromagnetic radiation propagating along the hollow core of the fiber, allowing photonic bandgap guidance of light to be implemented.[16,17] Owing to the high reflectivity of microstructure cladding, optical losses can be substantially reduced in such fibers, offering a unique opportunity of implementing non-linear optical interactions of waveguide modes with transverse sizes of several microns in a gas medium.[18] Such fibers, as recently shown by Benabid *et al*.,[19] allow the threshold of stimulated Raman scattering (SRS) in molecular hydrogen filling the fiber core to be considerably lowered, suggesting a means to improve considerably the efficiency of the SRS process.

The goal of this paper is to demonstrate that hollow-core photonic-crystal fibers (PCFs) provide an opportunity to enhance substantially FWM processes, including CARS-type interactions, thus suggesting new solutions for frequency conversion of high-intensity ultrashort laser pulses and offering recipes for improving the sensitivity of non-linear optical gas-phase analysis based on FWM and CARS techniques. We use 30 ps pulses of Nd : YAG fundamental radiation and its second harmonic to generate a signal at the frequency of the third harmonic through the FWM process $3\omega = 2\omega + 2\omega - \omega$. The efficiency achieved for this process in a 9 cm hollow-core PCF with a core diameter of about 13 μm will be shown to be ~800 times higher than the maximum FWM efficiency attainable with the same laser pulses in the tight-focusing regime.

EXPERIMENTAL

Our experiments were performed with hollow-core photonic-crystal fibers having an inner diameter of 13 μm and a period of the photonic-crystal cladding equal to 5 μm (Fig. 1). These microstructure fibers used in our experiments were fabricated[17] 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 photonic-crystal fibers. Transmission spectra of these hollow-core PCFs displayed characteristic well-pronounced isolated peaks [Fig. 2(a) and (b)], related to photonic bandgaps of the cladding.[16,17] Radiation with wavelengths lying away from photonic band gaps of the cladding leaks from the hollow core. Such leaky radiation modes are characterized by high losses, giving virtually no contribution to the signal at the output of the fiber.

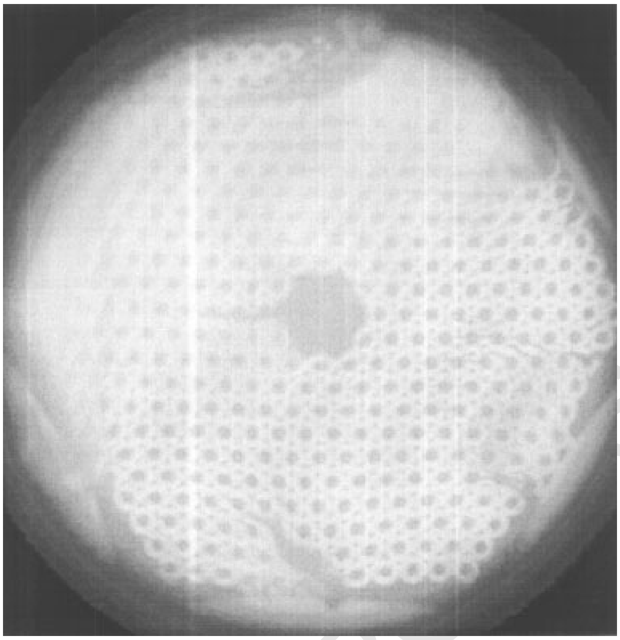

Figure 1. Cross-sectional image of a microstructure fiber with a two-dimensionally periodic cladding consisting of an array of identical capillaries. The hollow core of the fiber is formed by removing seven capillaries from the central part of the structure. The period of the structure in the cladding is ~5 μm and the core diameter is ~13 μm.

The spectra of air-guided modes in hollow photonic-crystal fibers can be tuned by changing the fiber cladding

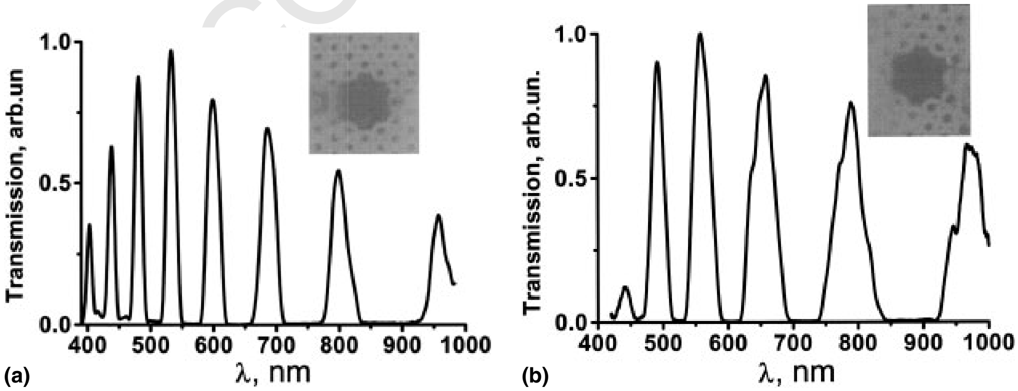

Figure 2. Transmission spectra measured for hollow-core photonic-crystal fibers with different cross-section geometries (shown in the insets). The period of the structure in the cladding is a ~5 μm and the core diameter is ~13 μm.

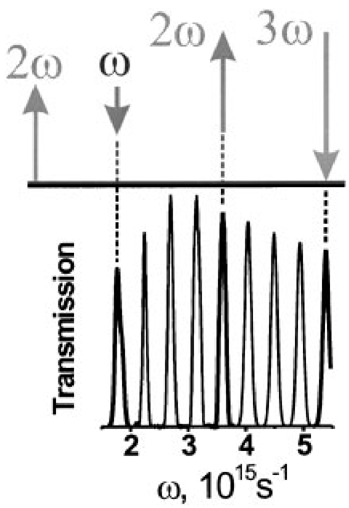

Figure 3. Diagram of the four-wave mixing process $3\omega = 2\omega + 2\omega - \omega$ (top) and the transmission spectrum of a hollow-core photonic-crystal fiber designed to transmit simultaneously the two-color pump (at 1.06 and 0.53 μm) and the FWM signal (bottom).

structure. In Fig. 2(a) and (b), this tunability option is illustrated by transmission spectra measured for hollow photonic-crystal fibers with different cross-section structures [shown in the insets to Fig. 2(a) and (b)]. The model of a coaxial Bragg waveguide[20] and a numerical analysis based on polynomial expansions of fields and two-dimensional refractive index profile in hollow-core PCFs[21,22] were used to design fibers with desirable transmission spectra. Hollow-core PCFs employed in our FWM experiments were designed in such a way as to provide maximum transmission simultaneously for the fundamental radiation of an Nd : YAG laser, and also for its second and third harmonics. The transmission spectrum of a hollow-core PCF designed for our FWM experiments is shown in the lower part of Fig. 3. Such a structure of the transmission spectrum is ideally suited for a non-linear optical interaction resulting in the generation of a signal at the frequency of the third harmonic through the $3\omega = 2\omega + 2\omega - \omega$ FWM process (the upper panel of Fig. 3). The magnitude of optical losses was estimated as 0.09 cm^{-1} at a wavelength of 1.06 μm and 0.08 cm^{-1} at 0.53 μm for these fibers. The FWM waveguide enhancement ratio μ under these conditions may be[23] as high as 800–1000.

Our experimental set-up was based on a picosecond laser system, which generated two-color pump radiation for the FWM process at the wavelengths of 1.06 and 0.53 μm. The picosecond laser included a passively mode-locked Nd : YAG master oscillator with negative-feedback-controlled cavity *Q*-factor,[24] a single-pulse selection unit and amplifying stages. Passive mode locking in the master oscillator was implemented with the use of a saturable absorber film, which was placed in front of the rear cavity mirror and which made it possible to generate laser pulses with a duration of about 30 ps. Negative feedback was introduced by inserting an electro-optical switch controlled with a fast-response photomultiplier inside the cavity. An electro-optical switch was used to separate a single pulse from this train. The energy of a single 30 ps laser pulse thus selected ranged from 30 to 40 μJ. The single-pulse selection unit also served as an optical decoupler, suppressing the parasitic feedback between amplifying stages and the master oscillator and preventing radiation reflected from optical elements of the amplification system from influencing the formation of pulse trains in the master oscillator.

An amplified single pulse of 1.06 μm radiation is used to generate the second harmonic in a DKDP crystal (Fig. 4). The second harmonic signal is separated from the fundamental beam with a beamsplitter. The fundamental and second-harmonic beams with energies of about 30 μJ are then coupled into a hollow-core PCF (Fig. 4) placed on a three-dimensional translation stage. The length of the hollow-core PCF was equal to 9 cm, slightly less than the length optimal for the FWM process, $l_{\text{opt}}^{\text{FWM}} = \ln 3/\alpha$.

RESULTS AND DISCUSSION

Linearly polarized fundamental and second-harmonic pulses of Nd : YAG laser radiation coupled into a 9 cm hollow PCF excited the fundamental waveguide modes of PCF fibers with a typical light intensity distribution shown in the inset

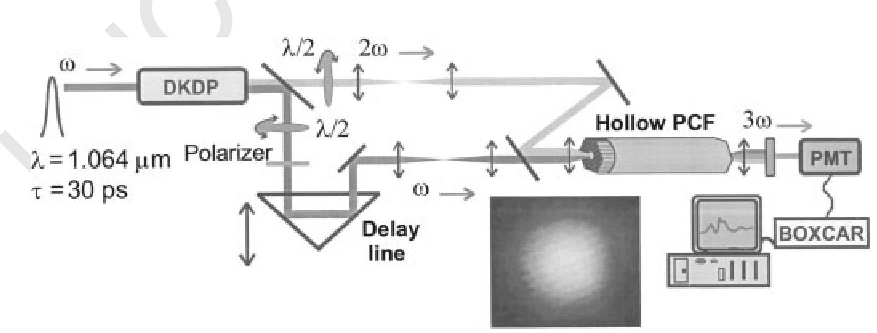

Figure 4. Experimental set-up for the investigation of FWM in a hollow-core photonic-crystal fiber: DKDP, crystal for second-harmonic generation; PMT, photomultiplier. The inset shows the transverse intensity distribution in the air-guided mode of the hollow-core photonic-crystal fiber whose cross-sectional view is shown in Fig. 1.

to Fig. 4. The signal at the frequency of the third harmonic of fundamental radiation, 3ω, 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 at least 70 times less efficient than two-color FWM, and the 3ω signal in two-color FWM experiments is nearly entirely generated through the difference-frequency generation process. This relation between the efficiencies of direct third-harmonic generation and the difference-frequency FWM process is apparently due to the difference in the phase mismatches corresponding to these processes. This hypothesis is supported by the results of our measurements[25] performed on the spectral phase of femtosecond pulses transmitted through a hollow-core PCF using the spectral interferometry for direct electric field reconstruction (SPIDER) technique.[26] According to these studies, the mismatch $|\Delta\beta_{\mathrm{FWM}}| = |2\beta_{2\omega} - \beta_{\omega} - \beta_{3\omega}|$ of propagation constants β_{ω}, $\beta_{2\omega}$ and $\beta_{3\omega}$ of waveguide modes at frequencies $\omega, 2\omega$ and 3ω, respectively, involved in the difference-frequency FWM process is much less than the propagation-constant mismatch $|\Delta\beta_{\mathrm{THG}}| = |\beta_{3\omega} - 3\beta_{\omega}|$ for direct third-harmonic generation. Among other $\chi^{(3)}$-related processes, sum-frequency generation $4\omega = 2\omega + \omega + \omega$ could have a non-negligible efficiency according to the spectral phase distribution under the conditions of our experiments. However, no waveguide mode was supported at the frequency of the fourth harmonic in the hollow core of our PCFs.

The energy of the FWM signal produced in a hollow-core PCF was compared with the energy of the FWM signal generated by tightly focused 1.06 and 0.53 μm pump beams with the same energies (the diagrams of the waveguide and tight-focusing regimes of FWM are shown in Fig. 5). This comparison allowed the FWM waveguide enhancement factor μ to be estimated as ~800. This result qualitatively agrees with our expectations based on the theoretical analysis of waveguide FWM in hollow PCFs.[23] With the validity of our expectations confirmed by experimental results, we can predict that even higher enhancement factors can be achieved for FWM processes with less lossy hollow-core photonic-crystal and microstructure fibers. In particular, the FWM waveguide enhancement factor may exceed four orders of magnitude in the case of hollow microstructure fibers with the magnitude of optical losses equal to 0.01 cm^{-1}.

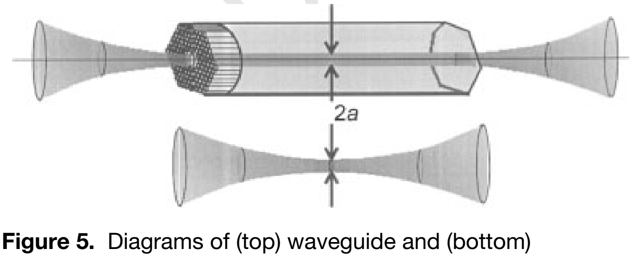

Figure 5. Diagrams of (top) waveguide and (bottom) tight-focusing regimes of FWM.

Table 1. Enhanced four-wave mixing $3\omega = 2\omega + 2\omega - \omega$ (ω is the frequency of fundamental radiation of an Nd : YAG laser) of 30 ps pulses in a solid-cladding hollow fiber (HF)[27] and a hollow-core photonic-crystal fiber (PCF)[a]

	a/μm	l/cm	μ	E_{ω}/μJ	$E_{2\omega}$/μJ
HF	100	10	15	10	10
PCF	13	9	800	2	2

[a] Notation: a, inner diameter of a hollow fiber; l, fiber length; μ, waveguide FWM enhancement; E_{ω} and $E_{2\omega}$, typical energies of the pump beams with frequencies ω and 2ω.

Table 1 compares the waveguide enhancement factor μ of the FWM process $3\omega = 2\omega + 2\omega - \omega$ attainable with a hollow-core PCF with the waveguide enhancement achieved for the same FWM process involving laser pulses with the same durations and wavelengths in experiments[27] performed with conventional, solid-cladding hollow fibers. This comparison shows that a 9 cm hollow-core PCF with an inner diameter of 13 μm designed in such a way as to provide simultaneously a high transmission for the two-color pump and the FWM signal allows the FWM enhancement factor to be increased by a factor of more than 50 compared with a 10-cm solid-cladding hollow fiber with an inner diameter of 100 μm. The FWM signal can be reliably detected under these conditions with micro- and even submicrojoule pump pulses, i.e. at the level of pump energies unprecedentedly low for non-resonant FWM in the gas phase.

To analyze the sensitivity of the FWM process in air-guided modes of a hollow-core PCF to the relative orientation of polarization vectors of input pump pulses, we performed FWM experiments with linear polarized fundamental and second-harmonic pump pulses having parallel and orthogonal polarization vectors. The intensity of the FWM signal generated by pump beams with parallel polarization vectors was 9–12 times higher than the intensity of the FWM signal produced by pump beams with the same intensities, but orthogonal polarizations. This ratio of the intensities of FWM signals produced by pump beams with linearly and orthogonal polarizations is, roughly, of the same order of magnitude as the ratio of non-resonant cubic susceptibilities responsible for FWM in parallel and orthogonal pump-beam polarization geometries, dictated by Kleinman relations,[28] $|\chi_{1221}|^2/|\chi_{1111}|^2 = 1/9$. This approximate, order of magnitude, conservation of Kleinman relations may imply, in particular, that orthogonal, quasi-linear polarization states, constituting the fundamental mode of the hollow-core PCF, may be selectively addressed with different polarization configurations of pump beams involved in the FWM process. Such a possibility would allow the whole family of FWM and CARS polarization techniques to be extended to waveguide regimes in photonic-crystal fibers.

CONCLUSION

Hollow-core photonic-crystal fibers providing maximum transmission simultaneously for a two-color pump (1.06 μm fundamental radiation from an Nd : YAG laser and its second harmonic) and the FWM signal (at the frequency of the third harmonic of Nd : YAG laser radiation) were designed to optimize the generation of the signal at the frequency of the third harmonic through an FWM process involving the fundamental and second-harmonic radiation of an Nd : YAG laser as a pump. These fibers substantially enhance the four-wave mixing of laser pulses in a gas filling the fiber core, offering a unique opportunity of improving the sensitivity of FWM and CARS spectroscopy by implementing non-linear optical interactions of waveguide modes with transverse sizes of several microns in a gas medium. The efficiency achieved for such a process with 30 ps pulses of Nd : YAG fundamental radiation and its second harmonic coupled into a 9 cm hollow-core photonic-crystal fiber with a core diameter of about 13 μm is ∼800 times higher than the maximum FWM efficiency attainable with the same laser pulses in the tight-focusing regime. A reliably detectable FWM signal was generated under these conditions with micro- and even submicrojoule picosecond pump pulses. The dependence of the FWM efficiency on the relative orientation of polarization vectors of input pump pulses suggests the extension of FWM and CARS polarization techniques to air-guided modes in photonic-crystal fibers.

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