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14. ABSTRACT

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**ENHANCED ULTRAFAST NONLINEAR OPTICS WITH
MICROSTRUCTURE FIBERS AND PHOTONIC CRYSTALS**

**Final Technical Report
by**

Aleksei Zheltikov

(July 2004)

United States Army

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ABSTRACT

A broad class of nonlinear-optical phenomena enhanced in microstructure and photonic-crystal fibers has been studied both experimentally and theoretically as a part of this project. Hollow-core photonic-crystal fibers capable of transporting sub-100-fs pulses of Ti: sapphire laser radiation in one of their transmission peaks centered around 800 nm have been designed and demonstrated. These fibers are shown to enhance self-phase modulation of submicrojoule 100-fs Ti: sapphire laser pulses, allowing a spectral bandwidth of 35 nm to be achieved with an 8-cm PCF sample. Two cascaded hollow-core photonic-crystal fibers with slightly shifted, but still overlapping transmission peaks are shown to function as an optical diode for ultrashort laser pulses. Submicrojoule 100-fs Ti: sapphire laser pulses with a spectrum falling within the passband of one of the fibers, but outside the passband of the second fiber, experience spectral broadening due to self-phase modulation in the first fiber. A part of this self-phase-modulation-broadened spectrum is then transmitted through the second fiber. Identical short pulses propagating in the opposite direction are blocked by the second fiber with a shifted passband. A forward-to-backward signal ratio exceeding 40 is achieved with the created photonic-crystal fiber diode for 0.9- μ J, 100-fs pulses of 800-nm Ti: sapphire laser radiation. Self-phase-modulation-induced spectral broadening of laser pulses in air-guided modes of hollow photonic-crystal fibers (PCFs) is shown to allow the creation of fiber-optic limiters for high-intensity ultrashort laser pulses. The performance of PCF limiters is analyzed in terms of elementary theory of self-phase modulation. Experiments performed with 100-fs microjoule pulses of 800-nm Ti: sapphire laser radiation demonstrate the potential of hollow PCFs as limiters for 10-MW ultrashort laser pulses and show the possibility to switch the limiting level of output radiation energy by guiding femtosecond pulses in different PCF modes. The total of 20 papers have been published in peer-reviewed international scientific journals as a result of this project. Ten invited talks have been given at major meetings, including one at the Conference on Lasers and Electro-Optics (CLEO2004) -- the leading forum in laser science.

List of keywords: photonic band gaps, photonic crystals, ultrashort pulses, waveguides, fibers, nonlinear optics, wave mixing

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Appendix III: Reprint of the paper S.O Konorov, V.P Mitrokhin, A.B Fedotov, D.A Sidorov-Biryukov, V.I. Beloglazov, N.B. Skibina, E. Wintner, M. Scalora, and A.M. Zheltikov, Hollow-core photonic-crystal fibres for laser dentistry, *Phys. Med. Biol.* 49, 1359 (2004)

Appendix IV: Reprint of the paper A.B. Fedotov, I. Bugar, D.A. Sidorov-Biryukov, E.E. Serebryannikov, D. Chorvat Jr., M. Scalora, D. Chorvat, and A.M. Zheltikov, Pump-depleting four-wave mixing in supercontinuum-generating microstructure fibers, *Applied Physics B*, 77, 313–317 (2003).

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Appendix VI: Reprint of the paper S. O. Konorov, E. E. Serebryannikov, A. M. Zheltikov, Ping Zhou, A.P. Tarasevitch, and D. von der Linde, Mode-Controlled Colors from Microstructure Fibers, *Optics Express*, 12, 730 (2004).

TECHNICAL REPORT

SELF-PHASE MODULATION OF SUBMICROJoule FEMTOSECOND PULSES IN A HOLLOW-CORE PHOTONIC-CRYSTAL FIBER

Hollow-core photonic-crystal fibers capable of transporting sub-100-fs pulses of Ti: sapphire laser radiation in one of their transmission peaks centered around 800 nm have been designed and demonstrated. These fibers are shown to enhance self-phase modulation of submicrojoule 100-fs Ti: sapphire laser pulses, allowing a spectral bandwidth of 35 nm to be achieved with an 8-cm PCF sample.

EXPERIMENTAL DEMONSTRATION OF A PHOTONIC-CRYSTAL-FIBER OPTICAL DIODE

Two cascaded hollow-core photonic-crystal fibers with slightly shifted, but still overlapping transmission peaks are shown to function as an optical diode for ultrashort laser pulses. Submicrojoule 100-fs Ti: sapphire laser pulses with a spectrum falling within the passband of one of the fibers, but outside the passband of the second fiber, experience spectral broadening due to self-phase modulation in the first fiber. A part of this self-phase-modulation-broadened spectrum is then transmitted through the second fiber. Identical short pulses propagating in the opposite direction are blocked by the second

fiber with a shifted passband. A forward-to-backward signal ratio exceeding 40 is achieved with the created photonic-crystal fiber diode for 0.9- μ J, 100-fs pulses of 800-nm Ti: sapphire laser radiation.

LIMITING OF MICROJOULE FEMTOSECOND PULSES IN AIR-GUIDED MODES OF A HOLLOW PHOTONIC-CRYSTAL FIBER

Self-phase-modulation-induced spectral broadening of laser pulses in air-guided modes of hollow photonic-crystal fibers (PCFs) is shown to allow the creation of fiber-optic limiters for high-intensity ultrashort laser pulses. The performance of PCF limiters is analyzed in terms of elementary theory of self-phase modulation. Experiments performed with 100-fs microjoule pulses of 800-nm Ti: sapphire laser radiation demonstrate the potential of hollow PCFs as limiters for 10-MW ultrashort laser pulses and show the possibility to switch the limiting level of output radiation energy by guiding femtosecond pulses in different PCF modes.

SWITCHING INTENSE LASER PULSES GUIDED BY KERR-EFFECT-MODIFIED MODES OF A HOLLOW-CORE PHOTONIC-CRYSTAL FIBER

A Kerr-nonlinearity-induced profile of the refractive index in the hollow core of a photonic-crystal fiber (PCF) changes the spectrum of propagation constants of air-guided modes, effectively shifting the passbands in fiber transmission, controlled by photonic band gaps (PBGs) of the cladding. This effect is shown to allow the creation of fiber switches for high-intensity laser pulses. The Kerr-nonlinearity control of air-guided modes in PCFs and the performance of a PCF switch are quantified by solving the propagation equation for the slowly varying envelope of a laser pulse guided in Kerr-effect-modified PCF modes. The spatial dynamics of the light field in a PBG waveguide switch is analyzed with the use of the slowly varying envelope approximation, demonstrating high contrasts of optical switching with PBG waveguides and hollow PCFs.

COHERENT ANTI-STOKES RAMAN SCATTERING IN ISOLATED AIR-GUIDED MODES OF A HOLLOW-CORE PHOTONIC-CRYSTAL FIBER

Hollow-core photonic-crystal fibers are shown to offer the unique possibility of coherent excitation and probing of Raman-active vibrations in molecules by isolated air-guided modes of electromagnetic radiation. A 3-cm section of a hollow photonic-crystal fiber is used to prepare isolated air-guided modes of pump and probe fields for a coherent excitation of 2331-cm⁻¹ Q-branch vibrations of molecular nitrogen in the gas filling the fiber core, enhancing coherent anti-Stokes Raman scattering through these vibrations by a factor of 15 relative to the regime of tight focusing.

SELF-CHANNELING OF SUBGIGAWATT FEMTOSECOND LASER PULSES IN THE GROUND-STATE WAVEGUIDE INDUCED IN THE HOLLOW CORE OF A PHOTONIC-CRYSTAL FIBER

Femtosecond laser pulses with powers below the blowup threshold for self-focused beams are shown to experience spatial self-action in hollow-core photonic-crystal fibers filled with argon, nitrogen, and atmospheric air. Regardless of the transverse field distribution at the input of the fiber, the output beam pattern in this regime tends to a circularly symmetric profile, corresponding to a ground-state waveguide induced by laser pulses inside a hollow fiber.

PHASE-MATCHED FOUR-WAVE MIXING OF SUB-100-TW/CM² FEMTOSECOND LASER PULSES IN ISOLATED AIR-GUIDED MODES OF A HOLLOW PHOTONIC-CRYSTAL FIBER

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 $3\omega = 2\omega + 2\omega$ - ω four-wave mixing of fundamental (ω) and second-harmonic (2ω) sub-100-TW/cm² 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.

SPECTRALLY AND TEMPORALLY ISOLATED RAMAN SOLITON FEATURES IN MICROSTRUCTURE FIBERS VISUALIZED BY CROSS-CORRELATION FREQUENCY-RESOLVED OPTICAL GATING

We study soliton phenomena accompanying the propagation of femtosecond Cr: forsterite-laser pulses through a microstructure fiber in the regime of efficient anti-Stokes frequency conversion. The dispersion of the fiber is designed in such a way as to minimize the group delay of the 1.25- μm pump and the Stokes pulse within the length of soliton pulse compression in the regime of anomalous dispersion. Spectrally and temporally isolated solitonic features, resulting from soliton self-frequency shift, are detected at the output of such a microstructure fiber by means of cross-correlation frequency-resolved optical gating.

CROSS-CORRELATION FREQUENCY-RESOLVED OPTICALLY GATED COHERENT ANTI-STOKES RAMAN SCATTERING WITH FREQUENCY-CONVERTING PHOTONIC-CRYSTAL FIBERS

Photonic-crystal fibers provide a high efficiency of frequency upconversion of regeneratively amplified femtosecond pulses of a Cr: forsterite laser, permitting the generation of subpicosecond anti-Stokes pulses with a smooth temporal envelope and a linear positive chirp, ideally suited for femtosecond coherent nonlinear spectroscopy. These pulses from a photonic-crystal fiber were cross-correlated in our experiments with the femtosecond second-harmonic output of the Cr: forsterite laser in toluene solution, used as a test object, in boxcars geometry to measure the spectra of coherent anti-Stokes Raman scattering (CARS) of toluene molecules (XFROG CARS).

LARGE-CORE-AREA HOLLOW PHOTONIC-CRYSTAL FIBERS

We demonstrate hollow-core photonic-crystal fibers (PCFs) with a core diameter of 45 μm and a period of the photonic-crystal cladding of 7 μm . These fibers are shown to allow a waveguide delivery of 4-mJ 15-ns pulses of 1.06- μm radiation of a Q-switched Nd: YAG laser. Hollow PCFs reported in this work help to bridge the gap between standard, solid-cladding hollow fibers and hollow PCFs in terms of effective guided-mode areas, offering attractive solutions for high-field physics, laser technologies, and ultrafast photonics.

ANTI-STOKES GENERATION IN GUIDED MODES OF PHOTONIC-CRYSTAL FIBERS MODIFIED WITH AN ARRAY OF NANOHOLES

Periodic arrays of air holes with diameters ranging from about 80 nm up to 400 nm are shown to modify intensity profiles of guided modes in photonic-crystal fibers, suggesting new ways of fiber dispersion engineering and modal birefringence control. Guided modes supported by such fibers provide a high efficiency of anti-Stokes frequency conversion for 100-fs pulses of a Cr: forsterite laser.

FREQUENCY-TUNABLE ANTI-STOKES LINE EMISSION BY EIGENMODES OF A BIREFRINGENT MICROSTRUCTURE FIBER

Birefringent microstructure fibers are shown to allow efficient generation of frequency-tunable anti-Stokes line emission as a result of nonlinear-optical spectral transformation of unamplified femtosecond Ti: sapphire laser pulses. Femtosecond pulses of 820-nm pump radiation polarized along the fast and slow axes of the elliptical core of the microstructure fiber generate intense blue-shifted lines centered at 490 and 510 nm, respectively, observed as bright blue and green emission at the output of a 10-cm microstructure fiber.

GENERATION OF FEMTOSECOND ANTI-STOKES PULSES THROUGH PHASE-MATCHED PARAMETRIC FOUR-WAVE MIXING IN A PHOTONIC-CRYSTAL FIBER

Phase-matched parametric four-wave mixing in higher order guided modes of a photonic-crystal fiber is shown to result in an efficient decay of 40-fs 800-nm Ti: sapphire-laser pump pulses into an anti-Stokes signal with a central wavelength around 590–600 nm and a Stokes signal centered at 1.25 μm . The photonic crystal fiber is designed in such a way as to minimize the group-velocity dispersion at the pump wavelength, phase-match the parametric four-wave mixing process, and reduce the group delay

between the pump and the anti-Stokes pulses. The duration of the anti-Stokes pulse under these conditions, as shown by cross-correlation frequency-resolved optical gating measurements, is less than 200 fs.

MODE-CONTROLLED COLORS FROM MICROSTRUCTURE FIBERS

We experimentally demonstrate mode-controlled spectral transformation of femtosecond laser pulses in microstructure fibers. Depending on the waveguide mode excited in the fiber, 30-fs Ti: sapphire laser pulses can either generate a broadband emission or produce isolated spectral components in the spectrum of output radiation. This method is used to tune the frequencies dominating the output spectra, controlled by phase matching for four-wave mixing processes.

HOLLOW PHOTONIC-CRYSTAL FIBERS FOR LASER DENTISTRY

Sequences of picosecond pulses of 1.06- μm Nd:YAG-laser radiation with a total energy of about 2 mJ are transmitted through a hollow-core photonic-crystal fiber with a core diameter of approximately 14 μm and are focused on a tooth surface *in vitro* to ablate dental tissue. The hollow-core photonic crystal fiber is shown to support the single-fundamental-mode regime for 1.06- μm laser radiation, serving as a spatial filter and allowing the laser beam quality to be substantially improved. The same fiber is used to transmit emission from plasmas produced by laser pulses on the tooth surface in the backward direction for detection and optical diagnostics.

NANOSCALE NONLINEAR OPTICS WITH POROUS MATERIALS

Our work was also aimed at creating random photonic structures for a control, frequency conversion, and spectral transformation of ultrashort pulses. Nano and mesoporous silicon, silicon dioxide, and gallium phosphide samples and silica aerogels were studied as a part of this work. Coherent anti-Stokes Raman scattering (CARS) is shown to allow a reliable detection of Raman-active modes of toluene molecules in solution and gas-phase nitrogen molecules infiltrated into mesoporous silica aerogels. Due to the interference nature of CARS spectra and the high spatial resolution, CARS proves to be ideally suited for local probing of nanostructures and nanocomposite materials, as well as for sensing of Raman-active species in the liquid and gas phase in a nanocomposite host, allowing a nano-scale extension of CARS methodology, referred to as nanoCARS.

THE LIST OF PARTICIPANTS OF THE PROJECT

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APPENDICES

Appendix I: Reprint of the paper M.S. Syrchin, A.M. Zheltikov, and M. Scalora, Analytical Treatment of Self-Phase Modulation Beyond the Slowly Varying Envelope Approximation, *Physical Review A*, **69**, 053803 (2004).

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Appendix III: Reprint of the paper S.O Konorov, V.P Mitrokhin, A.B Fedotov, D.A Sidorov-Biryukov, V.I. Beloglazov, N.B. Skibina, E. Wintner, M. Scalora, and A.M. Zheltikov, Hollow-core photonic-crystal fibres for laser dentistry, *Phys. Med. Biol.* **49**, 1359 (2004)

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