Final research report on project „OTKA NN 107235"

2012. nov. 1 – 2016. dec. 31.

The main purpose of the project was to optimize the production of XUV pulses in a high-order-harmonic generation scheme, in order to enable the realization of time-resolved molecular dynamics studies. During the four years of the project the main emphasis has shifted slightly. In this report I summarize the results obtained.

The works were performed in collaboration with our long-standing experimental partners (*Max Born Institute*, Berlin, Germany; *Lund University*, Sweden). During the project we have also started new collaborations with the *Max Planck Insitute for Quantum Optics*, Garching, Germany and the *Institute of Optics and Quantum Electronics*, Jena, Germany, and *Institut Lumière Matière*, Lyon, France.

(Our achievements have been presented in conferences, and published in per-reviewed journals. About each result I list the highest visibility publication. The participants of the OTKA project are marked by underlining their names.)

1) Numerical studies in order to optimize the XUV source

a) The effect of an assisting THz / MIR field on XUV generation by a laser pulse

We investigated how the presence of an intense THz field modifies the high harmonic generation process, leading to a significant increase of the cutoff photon energy or of the photon flux. This was the proposed scheme to be realized for production of the XUV radiation required for the main experiments of the project.

1. Emeric Balogh, József A. Fülöp, János Hebling, Péter Dombi, Győző Farkas, Katalin Varjú: *Application of high intensity THz pulses for gas high harmonic generation*, Central European Journal of Physics **11** (9), 1135-1140 (2013)

To improve the yield of high-order harmonic generation we studied macroscopic enhancement of the produced radiation in quasi-phase-matching arrangements. In the numerical study we analysed quasi-phase-matching induced by low-intensity assisting fields. We investigated the required assisting field-strength dependence on the wavelength and intensity of the driving field, harmonic order, trajectory class and period of the assisting field.

2. E. Balogh and K. Varju, *Quasi-phase-matched high-order harmonic generation by low-intensity* assisting fields: Field strength scaling and bandwidth, JOSA B 33 (2), 230-238 (2016)

It is well known that the harmonic generation process scales unfavourably with the generating laser wavelength, thus MIR laser sources – although providing a mean to reach higher cutoff photon energies – enable the production of lower photon yields. Inspired by the results of the two-colour experiments described above we modelled high-order harmonic generation by a MIR pulse in the presence of a weak perturbative assisting near infrared pulse. We have found that the yield of the highest energy photons can be increased by an order of magnitude in the whole spectral range up to 160 eV.

3. Katalin Kovacs, Balazs Major, Emeric Balogh, Valer Tosa, and Katalin Varju: *High efficiency single attosecond pulse generation with a long wavelength pulse assisted by a weak near infrared pulse*, IEEE Journal of Selected Topics in Quantum Electronics **21** (5), 8700207 (2015)

b) Higher laser powers for efficient XUV generation

The available laser power has grown so much in the last 10 years, that today the main question is how can we utilize these powerful lasers to generate the brightest attosecond pulses. As high harmonics are produced in a gas target, we need to consider the effect of the gas on the driving field. This topic has been investigated, and presented in conferences, and two research papers.

4. V. Tosa, K. Kovács, B. Major, E. Balogh, K. Varjú, *Propagation effects in highly ionised gas media*, Quantum Electronics **46** (4) 321 – 326 (2016)
5. Valer Toșa, Katalin Kovács, Daniel Ursescu, and Katalin Varjú, *Characteristics of Femtosecond Laser Pulses Propagating in Multiply Ionized Rare Gases*, submitted to Nuclear Instruments and Methods B

Simply using more powerful laser pulses does not guarantee the generation of powerful attosecond pulses. Scaling up high order harmonic generation for increasing driving laser powers in the low density target regime was investigated. We create favorable phase matching conditions by balancing the effects of Gouy phase shift, neutral and plasma dispersion, thus increasing laser powers scale up the geometric parameters (focal length, target length).

6. C. M. Heyl, H. Coudert-Alteirac, M. Miranda, M. Louisy, K. Kovacs, V. Tosa, E. Balogh, K. Varjú, A. L'Huillier, A. Couairon, and C. L. Arnold, *Scale-invariant nonlinear optics in gases,* Optica 3 (1), **75** (2016)

The different experimental setups for high-order harmonic generation (HHG) have a lot of degree of freedom, all making it available to tune the properties of the generated extreme-ultraviolet (XUV) radiation. It is especially important for high power laser systems to choose these parameters properly for the efficient usage of the available laser power. As an example, the way to optimize the parameters of the high-harmonic beams is an important question for the highly tunable future beamlines of the ELI-ALPS institute.

We used our three-dimensional nonadiabatic simulation model of the HHG process for optimization of the yield and beam properties of a future ELI-ALPS beamline. The multidimensional parameter scan involved scanning of the available laser pulse energies, beam sizes, cell positions, cell lengths and gas pressures. The whole study involved the simulation and evaluation of 1920 cases overall making it possible to optimize generation parameters in this 5-dimensional parameter space. During this study the same scaling principle was utilized that was developed in collaboration with foreign parties as a part of this project [6]. The different outputs of the simulation allow optimization for harmonic yield, harmonic spectrum or XUV beam parameters, like divergence or other spatial, spectral or temporal properties. The first investigations focused on the increase of the XUV radiation yield, showing that the low pressure and long interaction volume, suggested by previous studies [P. Rudawski et al., Rev. Sci. Instr. 84, 073103 (2013); J. Rothhardt et al., New. J. Phys. 16, 033022 (2014)] is favorable. The evaluation of the simulations also showed the different phase-matching regimes of different parameters regions. A more detailed examination involving other XUV properties has also been initiated. A similar simulation series for another beamline of the ELI-ALPS institute was also carried out, the full evaluation of the results has not been finished yet.

7. B. Major, P. Cs. Kőrös, K. Kovács, E. Balogh, C.M. Heyl, A. L'Huillier, V. Tosa, and K. Varjú, *Optimizing high harmonic generation in a multidimensional parameter space,* Poster 3.2, Magyar Fizikus Vándorgyűlés, 24th-27th August 2016, Szeged, Hungary
8. K. Kovács, B. Major, B. Manschwetus, E. Balogh, S. Maclot, L. Rading, P. Rudawski, C.M. Heyl, H. Coudert-Alteirac, B. Farkas, V. Tosa, P. Johnsson, A. L'Huillier, K. Varjú, *Macroscopic*

optimization of high harmonic generation for high power laser pulses, 5th International Conference on Attosecond Physics, Saint-Sauveur, Canada, July 6-10th, 2015. (manuscript in preparation)

c) Optimization of the attosecond pulse structure using light-field synthesizers

As the generation of high-order harmonics is a highly nonlinear process, it is very sensitive of the properties of the generating laser pulse. The extreme fine tuning of the laser electric field available in light field synthesizers opens new directions in attosecond physics. The high number of tunable degrees of freedom in such an arrangement is as disadvantageous as beneficial, with practical limitations on applicability. To test the applicability of such an arrangement for attosecond pulse control, we used numerical methods. We applied a genetic algorithm to optimize the input parameters of the light field synthesizer to produce predetermined attosecond pulse shapes. Other than the specific results obtained, through this examination we tested the applicability of the models we use in the simulations.

9. E. Balogh, B. Bódi, V. Tosa, E. Goulielmakis, P. Dombi, and K. Varju, *Genetic optimization of attosecond pulse generation in light-field synthesizers*, Physical Review **A 90**, 023855 (2014)
10. B. Bódi, E. Balogh, V. Tosa, E. Goulielmakis, K. Varjú, And P. Dombi, *Attosecond pulse generation with an optimization loop in a light-field-synthesizer*, Optics Express **24** (19) 21957 (2016)

2) Experimental studies to optimize the XUV source

a) Detailed study of the temporal characteristics in HHG

For the optimization of the high harmonic source enabling molecular dynamic studies targeted in the project, both the highest photon energy and the photon number need to be increased. High harmonic signal might be increased by increasing the number of interacting atoms (increasing pressure). To study the behaviour of high harmonics with increasing pressure we performed a series of experiments, and discussed the results using a 1D model accounting for the propagation and absorption of the laser and harmonic beams.

We have observed that by increasing the pressure the signal strength first increases, then reaches an optimal value and starts decreasing. The signal loss is more dominant in the higher harmonics. We also noted that the group delay of the attosecond pulses decreases with increasing pressure.

11. D. Kroon, D. Guenot, M. Kotur, E. Balogh, E. W. Larsen, C. M. Heyl, M. Miranda, M. Gisselbrecht, J. Mauritsson, P. Johnsson, K. Varju, A. L'Huillier, and C. L. Arnold, *Attosecond pulse walk-off in high-order harmonic generation*, Optics Letters **39**, 2218 (2014)

b) Manifesting the NIR + MIR arrangements

To perform the experiments proposed in the present project very high photon energies need to be generated. We propose to achieve this aim by generating high harmonics of an 800 nm laser pulse in the presence of a strong MIR-THz pulse. We performed pilot experiments at our collaborating partner (MBI, Berlin), and confirmed the pressure dependence of the signal noted in the previous study.

Extensive numerical modelling of the process enabled us to understand specific features of the generation mechanism. (1) by positioning the (high pressure) generating medium well before the geometrical focus, we achieve a balance between geometrical focusing and plasma defocusing, similar to guiding and an almost flat intensity profile is reached along the cell – which is very beneficial for phase matching the generated radiation. (2) Due to beating between the two generating wavelengths and transient phase matching produced by the rapid ionization of the medium on the rising edge of the pulses we can produce an isolated attosecond burst by the two 50-fs-long laser pulses (this is usually achieved only by few-cycle pulses).

12. Bernd Schutte, Paul Weber, Katalin Kovacs, Emeric Balogh, Balazs Major, Valer Tosa, Nguyen Xuan Truong, Marc J J Vrakking, Katalin Varju, Arnaud Rouzee: *Bright soft X-ray bursts gated by transient phase-matching in two-color high-order harmonic generation*, Optics Express **23,** 33947 (2015)

During the analysis we also observed, that the radiation generated in the two-color scheme show specific features as a function of delay between the two pulses. The physical origin of these features was identified using model calculations.

13. E. Balogh, K. Kovács, B. Major, A. Rouzée, S. Han, B. Schütte, P. Weber, M. J.J. Vrakking, V. Tosa, K. Varjú, Spectral shifts and asymmetries in two-color high-order harmonic generation, 5th International Conference on Attosecond Physics, Saint-Sauveur, Canada, July 6-10th, 2015. *(manuscript in preparation)*

c) HHG experiments in Szeged

To achieve a higher degree of flexibility for performing measurements, we upgraded the high harmonic generation setup in the TeWaTI laboratory at the University of Szeged.

To drive the HHG source, we used 25 fs long, 1.5 mJ pulses of a Ti:sapphire based chirped-pulse amplification laser system operating at 200 Hz in the TeWaTi lab at University of Szeged, Department of Optics and Quantum Electronics. We demonstrated the generation of high-order harmonics in Ar by focusing these pulses with a lens of 20 cm focal length into a 2 mm short, low pressure (ca. 10 mbar) gas cell. The produced XUV radiation was filtered spatially, while the generating beam and the lower harmonics were suppressed and the natural chirp of the generated radiation was compensated by using a 200 nm Al filter. This allowed us to create a train of short attosecond pulses.

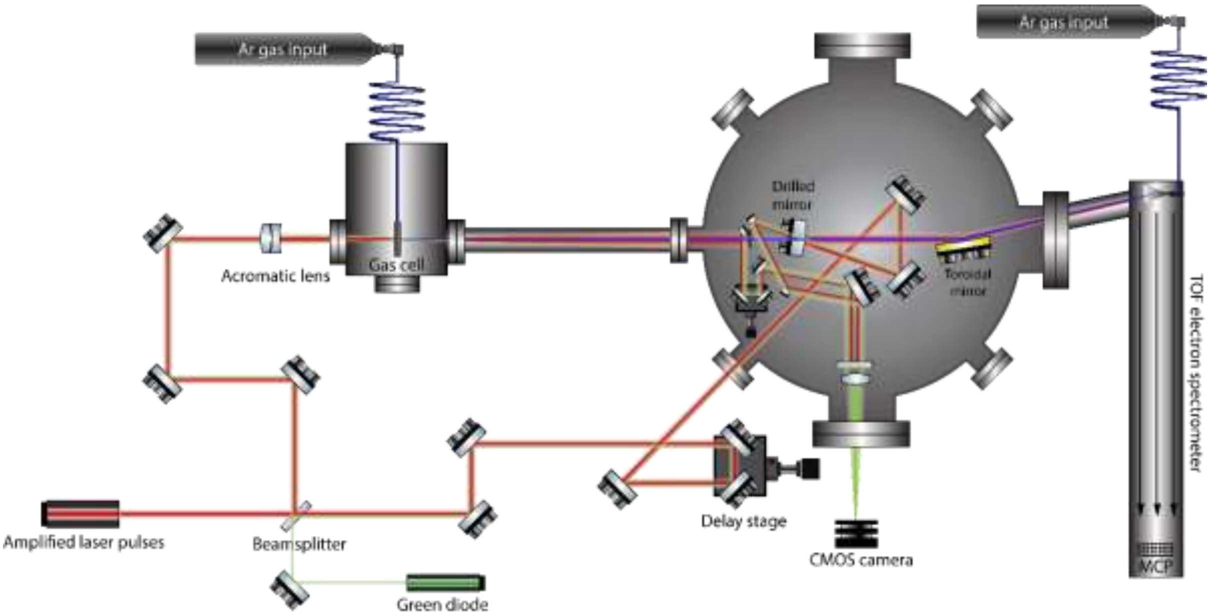

Fig. 1 Interferometric setup for temporal characterization of attosecond pulse trains

We used the RABBIT (Reconstruction of Attosecond Beating By Interference of Two-photon transitions) technique to characterize these pulses, for which the generating IR beam was split into two parts. The interferometer was stabilized by means of an auxiliary green beam (producing an interference pattern and providing a basis for the active stabilization), shown in Fig. 1. The temporal characterization verified the generation of 240 as pulses in a pulse train.

14. B. Nagyillés, B. Farkas, K. Varjú, *Attosecond beamline at the University of Szeged*, Magyar Fizikus Vándorgyűlés, Szeged 2016 (Poster)

3) Applications of the HHG source

a) Measuring photoemission delays

As described in the research proposal, the main reason for studying and optimising high order harmonic generation is to access the XUV – soft X-ray spectral range for light-matter interaction studies. We have not yet reached the several hundred eV regime targeted in the framework of the present project that will enable us to excite core electrons, but with the photon energies available we studied relative photoemission time delays between valence electrons in different noble gas atoms (Ar, Ne and He). The atoms were ionized by the high-harmonic attosecond pulse train synchronized with an infrared (IR) laser field. Photoemission delays were measured using an interferometric technique. The experimental results were compared with model calculations (Random Phase Approximation with Exchange and Multi-Congurational Hartree-Fock), and the influence of the different ionization angular channels were investigated.

15. Diego Guenot, David Kroon, Emeric Balogh, Esben Witting Larsen, Marija Kotur, Miguel Miranda, Thomas Fordell, Per Johnsson, Johan Mauritsson, Mathieu Gisselbrecht, Katalin Varju, Cord Arnold, Thomas Carette, Anatoly Kheifets, Eva Lindroth, Anne L'Huillier, and Marcus Dahlstrom, *Measurements of relative photoemission time delays in noble gas atoms*, J. Phys. B: At. Mol. Opt. Phys. **47**, 245602 (2014)

b) Experimental study of photodissociation of molecules

During an experimental campaign at Institut Lumière Matière, Lyon, France the goal was to study the photodissociation and photoionization dynamics of atoms and molecules. The unique high harmonic generation and laser setups of the laboratory make it possible to carry out XUV-pump UV-probe experiments and detect the result of the light-matter interaction using a velocity map imaging spectrometer (VMIS). The delay setup is adjustable both with femtosecond and sub-femtosecond steps, giving the possibility to follow both molecular- and electron dynamics induced by the incident photons. In the experiments carried out the photoionization properties of rare gases (helium, argon) were first studied using the "velocity map imaging" mode of the spectrometer for electron detection (see a VMI image of He ionized by the 17^{th} and 19^{th} harmonic of 800 nm in Fig.2(a)).

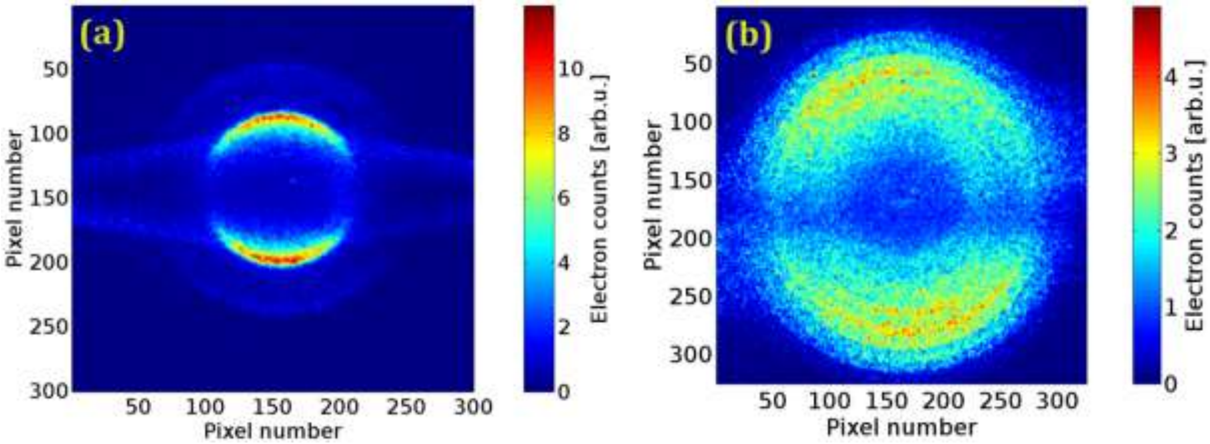

Fig.2. 2D momentum distribution of electrons resulting from the photoionization of (a) helium atoms and (b) nitrogen molecules by XUV pulse train generated in xenon atoms by HHG using 800 nm laser source.

The use of an UV probe having photons with energies equivalent to the energy spacing of the harmonics of the fundamental radiation gives a photoelectron signal corresponding to specific harmonic photon peaks, where each peak can result from the interference of electronic states going through different ionization channels. This allows for following the dynamics similarly to the basic idea of reconstruction of attosecond beating by interference of two-photon transitions (RABITT). The pump-probe measurements and the electron momentum distribution studies were also carried out on the aromatic hydrocarbon naphthalene ($C_{10}H_8$) and the diatomic molecule of the air constituent nitrogen (N_2), as it can be seen in the VMI image in Fig. 2 (b).

The time-of-flight spectrometer mode of the VMIS also allowed studying the photofragmentation of smaller molecules, like dimethylnitramine (DMNA, $C_2H_6N_2O_2$). In the performed experimental study the DMNA molecules, which were dissolved in gas phase, were irradiated by XUV pulse trains having different spectral properties generated using the HHG process. Then the time-of-flight mass spectrum of the resulted fragments was recorded, which involved pump-probe measurements with a UV probe. The obtained results are discussed in a manuscript, being prepared for publication.

c) Applications of the two-colour driven XUV source

Our collaborators at MBI have recently used the two-color high-order harmonic source, developed in the framework of this project, to generate a broadband XUV pulse extending from 40 eV to 90 eV to investigate the ultrafast changes of X-ray absorption in $LiBH_4$ polycristalline film near the Li K-edge (~60 eV) following a weak, non-resonant, 50 femtosecond, 800 nm laser excitation. Previous electron density maps derived from femtosecond X-ray diffraction experiment have shown that the laser electric field is responsible for a charge transfert from the $(BH_4)^-$ to neighboring Li^+ ions. It was demonstrated that the charge transfer lead to an additional atomic displacement along the Ag phonon Raman mode of the $LiBH_4$ thin film with a characteristic frequency around 10 THz.

4) Additional studies for HHG optimizations

a) HHG generation in solids

High-harmonic generation (HHG) in bulk solids can open new perspectives for the investigation of materials in a new regime of strong fields and attosecond timescales. Various experimental techniques related to high-order harmonic emission provides information for surface and bulk structure of solids. The arrangement can also act as a source of attosecond pulses.

The observation of HHG from atomically thin semiconductors demonstrates the ability to modulate the electronic properties of monolayer samples. Quantum mechanics was constructed to describe, for instance, the mechanisms giving rise to high harmonics emission in solids. Solving the eigenvalue problem for the periodic one-dimensional (1D) chain, the time-dependent Schrödinger equation (TDSE) can offer an obvious procedure for calculating the electron dynamics affected by laser-matter interaction. Practically, this means calculating the matrix elements of the perturbing Hamiltonian between the Bloch-states. We note that in our model even a local excitation can be taken into account (using e.g., position-dependent vector potential), but according to our experience, dipole approximation works very well for focal spot sizes of a few microns and above. The off-diagonal matrix elements reveal the interband transition, while the diagonal ones correspond to intraband motion.

The role of multiple conduction bands can also be investigated this way. The populations of the various bands in this figure are normalized to the initial population of the valence band, i.e., at t = 0, when the valence band is completely populated.

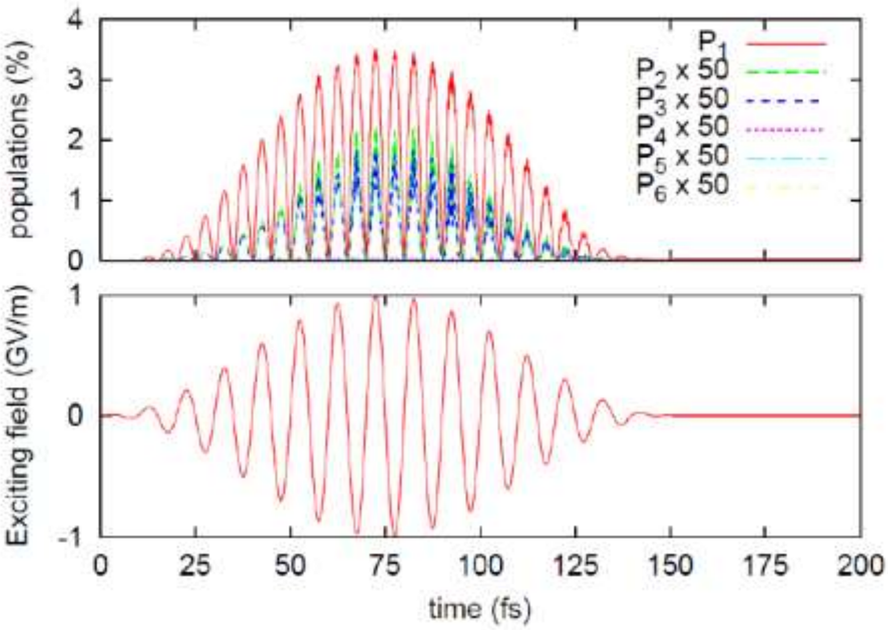

Figure 3: The population of the first six conduction bands as a function of time as shown in the upper panel. The waveform of the exciting pulse is depicted in the bottom panel. The peak value of the laser field is 1 GV/m, and the central wavelength is 3000 nm. For these parameters, it is clearly the first conduction band that plays the most important role.

An additional, important effect is related to the various decay mechanisms, that can be relevant already on the femtosecond timescale, but get even more significance for longer pulses. We can also include these effects, by considering quantum mechanical density matrices instead of pure states. The simplest example, related to pure dephasing, where interband coherences are assumed to decay. Although this is a purely phenomenological approach, the qualitative difference in the time evolution of the populations is clearly visible. Clearly, on a longer time scale, not only the coherences, but also the populations decay, and the system asymptotically reaches the ground state again.

During our work, we developed numerical codes (in C) to study HHG in bulk solids having crystal structure. The results have been presented in conferences, and is being written up in form of research papers.

16. P. Földi, V. Szaszkó-Bogár, K. Varjú, *Generation of high-order harmonics in solids: a theoretical model* (poster pres.), 4th ELI-ALPS User Workshop, Szeged (2016).
17. V. Szaszkó-Bogár, K. Varjú, P. Földi, *High-order harmonic genaration in bulk solids: the role of two-color excitation* (manuscript in preparation)

b) Measurements of waveguide dispersion

From their first discovery photonic crystal fibers (PCFs) acquired respectable attention due to their unique dispersion, birefringent, nonlinear and other guiding characteristics, which can be tailored by the proper design of their geometrical structure. The feasibility of guidance in air, i.e. in a hollow fiber core is especially advantageous as it facilitates high optical power transmission of the orders of 10^{14} W/cm^2 demonstrated in HC-800 type fibers (A. A. Ishaaya, et al., Opt. Express 17, 18630 (2009)) with remarkably low losses and reduced nonlinearity. These properties make hollow-core (HC) PCFs potential candidates for investigating intense light-matter interactions when the core is filled with a gaseous medium. Gas-filled HC-PCFs are also successfully employed for high harmonic generation (HHG) (O.H. Heckl et al., Appl. Phys. B 97, 369 (2009); T. Popmintchev et al., Nat. Photon. 4, 822 (2010)) with quite low HHG thresholds, easily obtainable using ultrafast diode-pumped solid-state oscillators. Additionally, HC-PCFs offer long effective interaction length, small mode areas and exceptional beam quality, therefore it is expected that the efficiency can be significantly enhanced by guiding and improved phase-matching using optimized dispersion engineering in PCFs.

Although fiber drawing methods have become quite accurate, the manufactured fiber structure usually slightly differs from the designed one. Even these small variations in the fiber geometry can result in rather different optical properties of the real fibers. Accordingly, experimental dispersion retrieval is of great interest to determine whether the fiber can be used for the application in question. Spectrally resolved interferometry is a widely used dispersion retrieval technique also applicable for fibers. In our research the goal was to find a method to measure the polarization dependent chromatic dispersion of a commercially available HC-800-02 PCF in different fiber placements as sometimes the fiber is bent or coiled during use. We also tested the additivity of the dispersion to find out whether the properties of the fiber can be extrapolated from testing a short sample, since in HHG fibers as short as a couple of mm are used. Using the Fourier-transform method we found that the dispersion properties of the tested fiber are different along orthogonal directions, however the dispersion was mostly independent on the placement and fiber length. The setup used in these measurements can be easily modified to measure different types of fibers.

18. Tímea Grósz, Attila P. Kovács, Katalin Varjú, *Unambiguous chromatic dispersion measurement along both polarization directions of a birefringent hollow-core photonic crystal fiber using spectral interferometry* submitted to Applied Optics