Characterization of high harmonic generation from gas under loose focusing condition

PhD thesis

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Introduction

With the invention of the laser [1], it became possible to generate coherent light pulses short enough to capture transient phenomena in matter. As laser technology evolved [2], the relentless pursuit of ever shorter pulse durations was driven by the principle that faster pulses unveil faster and more localized dynamics [3, 4]. This quest has not only deepened our understanding of ultrafast processes but has also catalyzed progress in applied research [5] and spurred innovations in industrial domains where precise timing and ultrafast control are paramount [6].

Despite these advances, there are fundamental limits on how short pulses can be directly generated from conventional laser gain media. These limits are imposed by the finite spectral bandwidth supported by the medium and by the difficulty of achieving uniform and controllable spectral phase across broad bandwidths. For state-of-the-art systems such as Ti:sapphire lasers and optical parametric amplifiers, the minimum pulse durations are typically in the 5–6 fs range [7, 8]. Pushing further into the single-cycle regime, near 2 fs in the NIR, requires post-compression techniques. These involve spectral broadening in hollow-core fibers [9–12] followed by dispersion compensation using chirped mirrors or acousto-optic modulators. While effective, these methods face practical limitations, beyond which direct laser-driven generation of shorter pulses becomes increasingly challenging. More recent developments include multi-pass cells [13, 14] and bulk-material compression schemes [15, 16].

To generate even shorter pulse durations, in the sub-femtosecond regime, one needs to go to shorter wavelength range so that enough bandwidth can be supported. To this end, alternative methods have been developed. Among them, high-order harmonic generation (HHG) has emerged as one of the most mature and experimentally accessible approach [17, 18]. Other concepts, such as ultrabroadband light synthesis with coherent phase control [19] or free-electron lasers (FELs) driven by plasma wakefield accelerators [20], promise extreme photon energies and ultrashort durations but typically demand large-scale infrastructure and complex instrumentation [21]. In contrast, HHG can be realized on a table-top scale while delivering femtosecond-to-attosecond pulses with high spatial and temporal coherence [22–24].

HHG is an inherently nonlinear process in which a strong laser field drives atoms or molecules to emit photons at multiples of the fundamental frequency, either odd or even depending on the symmetry properties of the medium [25] or electric field [26, 27], where generation takes place. The resulting spectrum can extend from the extreme ultraviolet (XUV) into the soft X-ray regime [28–30]. A breakthrough in the early 2000s showed that HHG could be harnessed to generate isolated attosecond bursts [23] or pulse trains [22], establishing the foundations of attosecond science and enabling direct time-domain studies of electron motion and ultrafast quantum dynamics.

The first experimental observation of HHG was reported in 1987 by independent groups in Chicago [31] and at CEA Saclay [32]. These early experiments used rare noble gases such as argon, krypton, and xenon, driven by the most advanced lasers of the time: Nd:YAG systems at 1064 nm [32], KrF excimer lasers at 248 nm [33, 34], and Ti:sapphire lasers at 800 nm [34–36]. Through the 1990s and early 2000s, the field expanded to include molecular targets, revealing the role of molecular orbitals and nuclear motion in strong-field dynamics. Their unique ability to resolve electronic, vibrational, and structural dynamics in real time has revolutionized the study of fundamental processes in atoms [31], molecules [37–39], and condensed matter systems [40–42]. Major milestones followed, including the demonstration HHG from plasma surfaces at ultra-high intensities [43–45], and the discovery of high-harmonic emission from bulk crystals in 2010 [46].

The solid-state breakthrough marked the beginning of a new chapter, emphasizing the potential of condensed matter systems as compact and efficient sources of attosecond light. Compared to gases, solids offer higher target densities and better integration prospects with photonic technologies, though the physics involves richer dynamics such as interband transitions and band-structure effects.

Today, HHG serves both as a powerful light source and as a precision probe, enabling applications ranging from ultrafast spectroscopy [22, 23, 47, 48] to coherent diffractive imaging [49, 50] to relativistic plasma optics [51–53] and time-resolved electron dynamics. Its continuing development remains central to advancing attosecond metrology and next-generation ultrafast light sources. The award of the 2023 Nobel Prize in Physics to Pierre Agostini, Ferenc Krausz, and Anne L'Huillier reflects not only the maturity of the field but also the importance of their pioneering contributions [54–56]. During the same period, laser technology has advanced at an impressive pace [57]. High–average-power sources with few-cycle pulse durations are now accessible [58], opening the door to new opportunities while also introducing serious challenges. Achieving efficient HHG with such lasers requires more than incremental refinements; it calls for expanded approaches to metrology and diagnostics.

Aims

In this thesis, I present several attosecond beamlines built around advanced laser systems, focusing on both on their technical realization and the scientific opportunities they make possible.

The central aim of this work was to develop and apply characterization techniques for different aspects of HHG. Using multi–tens of millijoule laser drivers, two distinct experimental approaches to achieve optimal phase matching had been carried out. One approach relied on short interaction lengths, such as gas jets, while the other used extended gas cells. As part of a user experiment, The aim was to investigate these two regimes systematically, comparing their performance under realistic conditions. [T1]

With the recent availability of multi–tens of millijoule lasers operating at 1 kHz repetition rate, such as the SYLOS2 system, HHG beamlines can now be driven at conditions that are highly attractive for applications requiring statistical averaging. At the same time, this repetition rate remains compatible with pulsed jet targets, which can serve both as generation media and as end-station targets. A new type of high repetition rate gas valve, promising high gas densities in the multi-kHz range, has recently emerged, though its characterization is not straightforward. The aim was to carry out detailed simulations and experiments to characterize this source with high spatial and temporal resolution, providing insight into its performance and limitations [T2, T3].

When the laser pulse duration approaches fewer than two optical cycles, the carrier-envelope phase becomes a critical parameter for HHG. One of my aims was to develop a method to estimate the CEP directly from measured harmonic spectra using machine learning. Through simulations of gas harmonics driven by the SYLOS laser, this technique feasibility can be evaluated under experimental conditions.

Solid state targets offer the possibility of probing material properties while generating broadband harmonic radiation from the infrared into the ultraviolet. At ELI ALPS, the MIR laser delivers pulses of only 1.5 optical cycles, which, when combined with a suitable crystal, allow strong CEP sensitivity to emerge. My goal was to measure this CEP dependence and to investigate whether machine learning methods could again be applied for CEP estimation, drawing a parallel to the gas-phase case [T4].

Results

T1 Part of the results presented were obtained during a user experiment investigating the dependence of high-order harmonic generation on medium length and gas pressure. For this experiment, I prepared the beamline and ensured that all subsystems were operating under optimal conditions. I characterized both the temporal and spatial properties of the driving infrared pulse and measured the characteristics of the generated XUV radiation. I optimized the generation conditions for the selected gas targets by adjusting the alignment, focusing geometry, and gas pressure. I also assembled, installed, and configured the gas cells, making sure they were suitable for the required medium lengths and buffer gas conditions. As part of the preparation, I performed preliminary measurements to verify the stability and performance of the setup. During the experimental campaign, I actively assisted the users with their measurements and supported them in analyzing and interpreting the recorded data.

The results confirm that there is an equivalence in terms of HHG yield between a short high pressure and a long low pressure medium. This predicted hyperbolic behavior of the medium length-pressure dependence had been confirmed. The model is not too sensitive to the laser intensity, which also confirmed by the measurements, since without it, it would have been difficult to tackle the issue with the energy loss due to the coupling of the laser beam into the cell between the different experimental cells. The difference between the XUV beamprofile in different medium length was also confirmed.

These findings are important during the designing phase of a new HHG beamline. When the XUV beam profile is not important as the high flux of photons, short high pressure jets has the advantage. Also, these are easier to implement compared to the generation in gas cell to loose focusing condition, and it can be the better choice when the laser energy is lower. The advantage of using long gas cells is that the XUV beam quality is better, and it is less sensitive to the pressure changes in the gas jets, and it is easier to implement a stable generation medium. This result had been published at [P3].

T2 I performed a complete spatial and temporal characterization of cantilever piezo gas valves, which are used for high-harmonic generation and spectroscopy experiments in both the SYLOS GHHG Long and SYLOS GHHG Compact beamlines. To accomplish this, I designed and built a stand-alone gas characterization end station that allowed precise measurements independent of the main beamline operation. I simulated interferometric fringe patterns corresponding to the expected gas density distributions and developed a dedicated data analysis code to retrieve accurate atomic number density profiles in the interaction region. I validated this approach against experimental data and demonstrated its reliability across a wide range of operating conditions. In addition, I designed the system to be flexible and adaptable to different gas delivery configurations, such as slit nozzles or multiple gas jets arranged in series, thereby extending its usefulness for future experimental setups and user needs. The results of this work were published in [P1].

T3 I conducted a series of measurements with high-harmonic generation to record the HHG yield fluctuation caused by gas density changes. For the HHG, I performed simulations using the gas density results recorded with the stand-alone gas characterization end station. The simulated results showed excellent agreement with the measured data, thereby confirming the accuracy and reliability of the density retrieval method. In parallel, I investigated light emitted by the generated plasma as a diagnostic tool to monitor potential variations in gas pressure during extended operation. This approach provided a practical, real-time monitoring tool, enabling the detection of gradual drifts or fluctuations in the gas delivery system without interrupting the experiment. These experimental result is presented in [P1].

T4 To investigate whether machine learning can be used to predict the carrier-envelope phase of a laser source from high-harmonic generation spectra, I carried out simulations of gas HHG, under conditions matching those of the SYLOS GHHG Long beamline. I trained a machine learning model using pairs of harmonic spectra and their corresponding CEP values. Finally, I tested the model on spectra it had not seen during training. The predictions reproduced the correct CEP values with only a small error, which confirms the validity of the approach.

I designed and constructed a dedicated experimental setup and I carried out the complete measurement campaign, which included aligning the beamline, optimizing the HHG process for stable spectral acquisition, and recording a large dataset under controlled experimental conditions. I subsequently performed a detailed analysis of the collected spectra, applying machine learning algorithms to establish the correlation between the spectral features and the CEP of the driving laser pulse. The results demonstrated that this approach is viable, offering the possibility of deploying it as an online diagnostic tool for CEP monitoring. Given its robustness, the method has the potential to be scaled and implemented in larger and more complex HHG beamlines, such as the SYLOS GHHG Long beamline, where real-time CEP tracking could significantly improve experimental stability and reproducibility. This experimental result is presented in [P2].

Publications

Own publications

(P1) **B. Nagyillés**, Z. Diveki, A. Nayak, M. Dumergue, B. Major, K. Varjú, and S. Kahaly, "Time-resolved investigation of a high-repetition-rate gas-jet target for high-harmonic generation," *Physical Review Applied*, vol. 20, no. 5, p. 054 048, 2023, MTMT: 34434071, IF: 4.4

(P2) **B. Nagyillés**, G. N. Nagy, B. Kiss, E. Cormier, P. Földi, K. Varjú, S. Kahaly, M. U. Kahaly, and Z. Diveki, "Mir laser cep estimation using machine learning concepts in bulk high harmonic generation," *Optics Express*, vol. 32, no. 26, p. 46 500, 2024, MTMT: 35670820, IF: 3.3

(P3) E. Appi, R. Weissenbilder, **B. Nagyillés**, Z. Diveki, J. Peschel, B. Farkas, M. Plach, F. Vismarra, V. Poulain, N. Weber, C. L. Arnold, K. Varjú, S. Kahaly, P. Eng-Johnsson, and A. L'Huillier, "Two phase-matching regimes in high-order harmonic generation," *Optics Express*, vol. 31, no. 20, p. 31 687, 2023, MTMT: 34393336, IF: 3.3

Other own publications

(O1) P. Ye, T. Csizmadia, L. G. Oldal, H. N. Gopalakrishna, M. Füle, Z. Filus, **B. Nagyillés**, Z. Divéki, T. Grósz, M. Dumergue, P. Jójárt, I. Seres, Z. Bengery, V. Zuba, Z. Várallyay, B. Major, F. Frassetto, M. Devetta, G. D. Lucarelli, M. Lucchini, B. Moio, S. Stagira, C. Vozzi, L. Poletto, M. Nisoli, D. Charalambidis, S. Kahaly, A. Zaïr, and K. Varjú, "Attosecond pulse generation at eli-alps 100 khz repetition rate beamline," *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 53, no. 15, p. 154 004, 2020, MTMT: 31421355, IF: 1.5

(O2) K. Chordiya, V. Despré, **B. Nagyillés**, F. Zeller, Z. Diveki, A. I. Kuleff, and M. U. Kahaly, "Photo-ionization initiated differential ultrafast charge migration: Impacts of molecular symmetries and tautomeric forms," *Physical Chemistry Chemical Physics*, vol. 25, no. 6, pp. 4472–4480, 2023, MTMT: 33107909, IF: 3.676

(O3) M. Staněk, O. Hort, L. Jurkovičová, M. Albrecht, O. Finke, **B. Nagyillés**, B. Farkas, T. Csizmadia, T. Grósz, A. Körmöczi, Z. Divéki, and J. Nejdl, "Photoelectric charge from metallic filters: An online xuv pulse energy diagnostics," *Applied Physics Letters*, vol. 125, no. 9, 2024, MTMT: 35256432, IF: 3.6

(O4) M. Shirozhan, S. Mondal, T. Grósz, **B. Nagyillés**, B. Farkas, A. Nayak, N. Ahmed, I. Dey, S. C. De Marco, K. Nelissen, M. Kiss, L. G. Oldal, T. Csizmadia, Z. Filus, M. De Marco, S. Madas, M. U. Kahaly, D. Charalambidis, P. Tzallas, E. Appi, R. Weissenbilder, P. Eng-Johnsson, A. L'Huillier, Z. Diveki, B. Major, K. Varjú, and S. Kahaly, "High-repetition-rate attosecond extreme ultraviolet beamlines at eli alps for studying ultrafast phenomena," *Ultrafast Science*, vol. 4, 2024, MTMT: 35294705, IF: 9.9

(O5) E. Skantzakis, I. Orfanos, A. Nayak, I. Makos, I. Liontos, E. Vassakis, T. Lamprou, V. Tsafas, T. Csizmadia, Z. Diveki, **B. Nagyillés**, B. Farkas, S. Mukhopadhyay, D. Rajak, S. Madas, M. Upadhyay Kahaly, S. Kahaly, R. Weissenbilder, P. Eng-Johnsson, E. Appi, A. L'Huillier, G. Sansone, K. Varju, L. A. A. Nikolopoulos, A. Emmanouilidou, P. Tzallas, and D. Charalambidis, "Nonlinear extreme ultraviolet applications with attosecond pulses," in *Progress in Ultrafast Intense Laser Science XVII*. Springer Nature Switzerland, 2024, pp. 1–24, MTMT: 35061801

Presentations, Posters, Conference papers

(C1) B. Major, B. Farkas, M. Dumergue, K. Kovacs, S. Kuehn, A. L'Huillier, **B. Nagyillés**, P. Rudawski, V. Tosa, P. Tzallas, D. Charalambidis, K. Osvay, G. Sansone, and K. Varjú, "The eli alps research infrastructure: Scaling attosecond pulse generation for a large scale infrastructure," in *High-Brightness Sources and Light-driven Interactions*, ser. HILAS, OSA, 2018, HW4A.1, MTMT: 27462331

(C2) **B. Nagyillés** et al., *Eli-alps sylos ghhg beamlines: Optimizing generation conditions for high-power laser pulses*, Poster at Atto VII, Szeged, Hungary, Demonstration of Attosecond beamline, 2019

(C3) **B. Nagyillés** et al., *High flux high harmonic generation at the sylos long beamline at eli-alps*, Poster at Atto VIII, Orlando, Florida, 2022

(C4) **B. Nagyillés** et al., *Time resolved characterization of high repetition rate gas jet target for high harmonic generation*, Poster at Atto IX, Jeju, Korea, 2023

(C5) R. Weissenbilder, E. Appi, J. Peschel, **B. Nagyillés**, S. Carlström, Z. Divéki, B. Farkas, M. Plach, K. Varjú, S. Kahaly, C. L. Arnold, P. Eng-Johnsson, and A. L'Huillier, "Hyperbolic trend in optimization of high-order harmonic generation in gases," in *2023 Conference on Lasers and Electro-Optics (CLEO)*, 2023, pp. 1–2, MTMT: 34434143

(C6) **B. Nagyillés** et al., *Mir laser cep estimation using machine learning concepts in bulk high harmonic generation*, Poster at 5th Attochem Workshop, Tenerife, Spain, 2024

(C7) **B. Nagyillés**, *Mir laser cep estimation using machine learning concepts in bulk high harmonic generation*, Talk at USTS 2024, Sitges, Spain, 2024

(C8) **B. Nagyillés**, *Extreme ultraviolet time-resolved photoelectron spectroscopy: Toward the ultimate probe of molecular dynamics*, Poster at XUVTRPES Workshop 2025, Freiburg, 2025

(C9) **B. Nagyillés**, *The effect of laser pulse duration on the yield of high harmonic generation*, Poster presentation at Atto X, Lund, Sweden, 2025

(C10) D. István, C. János, T. Szabolcs, T. L. Tamás, S. Tamás, G. P. Prasannan, A. D. Jenő, K. Balázs, D. Zsolt, **B. Nagyillés**, F. Balázs, K. Andor, and B. Ádám, *A sylos2a lézerrendszer és a vákuum szerepe a hullámfront stabilizálásában*, Kvantumelektronika : Szimpózium a hazai kvantumelektronikai kutatások eredményeiről Bibliogr.: 23. p. ; ill. 2025, MTMT: 36329464

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