Complete characterization of plasma mirrors and development of a single-shot carrier- envelope phase meter

Summary of PhD thesis

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2009

1. Preliminaries

With the invention of chirped pulse amplification the generation and amplification of ultrashort laser pulses to petawatt peak powers became possible. Such high-power pulses provide a unique means to investigate matter at extreme conditions. By focusing them into a tiny small spot, peak intensities of 10^{22} W/cm^2 can be generated that can instantaneously transform the surface of any solid target into a hot overdense plasma. At such extreme intensities and target temperatures relativistic interactions come to the fore opening the door to the exploration of a wide range of new phenomena. Experiments theoretically predicted for a long, like high-order harmonics generation from oscillating plasma surfaces, or proton acceleration from thin foil films are now routinely performed in many laboratories. The prerequisite to conduct laser-plasma experiments this kind is to ensure a clean interaction between the exposed solid target and the laser pulse. This means that prior to the arrival of the main laser pulse, radiation with a considerable intensity mustn't expose the target as it can alter the interaction. Unfortunately for technical reasons in the amplification process pedestals and leading prepulses are unavoidably generated. Their focused intensity is only a few orders of magnitude lower than that of the main pulse and thus it is well beyond the damage threshold of any target material. As a result prepulses and pedestal that overtake the main pulse generate a low density preplasma, which expands on the target surface and in place of the steep density gradient solid target the main pulse interacts with the low density preplasma. This unwanted phenomenon has existed since the invention of CPA lasers and has remained the main impediment to study laser-solid interaction at relativistic intensities.

Intensity contrast is the quantity that is used to characterize the temporal cleanness of laser pulses. It is the ratio of the intensity of the main pulse to that of the pedestal. Continuous efforts have been taken to improve the intensity contrast of high-power lasers by several optical methods and by incorporating all effective techniques into a laser system the contrast nowadays approaches 10^8 at best. This is several orders below the desired contrast ratios, as prepulses and the pedestal with an intensity above 10^7 W/cm^2 easily alter the interaction and lasers with focused intensities beyond 10^{20} W/cm^2 are now commonplace in many laser laboratories. Moreover with continuous developments available peak intensities are steadily increasing, while available laser contrast have been at the same level for more than a decade now. This shows that improving the laser contrast is one of the foremost challenges in high-intensity laser physics.

Plasma mirror (PM) which is an ultrafast self-induced optical shutter was proposed as a contrast improvement technique already in the early nineties. Its operation is based on the ultrafast ionization occurring at high intensities: a laser pulse is focused onto a transparent bulk target, and while the low intensity prepulses and pedestal mainly traverses the target and gets reflected only with the low Fresnel reflectivity, the rising edge of the main pulse with its high intensity generates a highly reflective flat plasma layer. The main pulse cleaned from the pedestal and prepulses is specularly reflected off the plasma layer thus the contrast of the reflected beam is significantly enhanced.

Although PM has been proposed for long as contrast improvement technique so far only proof-of principle studies have been conducted but no thorough characterization or practical implementation of this technique have been reported yet. My motivation in my Phd studies was: to demonstrate that PM can effectively improve the contrast of high-power lasers, to perform a complete experimental characterization of the PM, and to improve the temporal contrast of the 100 TW laser with the implementation of a double plasma mirror (DPM) setup and completely characterize its operation.

While the first part of my thesis focuses on the improvement of the temporal contrast of sub-picosecond high-power laser pulses – as it has been briefly presented above – the second part of my thesis deals with a fairly different topic, with the development of a single-shot carrier-envelope phase meter.

Thanks to the vast progress in ultrafast laser technology generation of amplified laser pulses comprising merely a few oscillation cycles of the electromagnetic field became possible to the turn of the millennium. The important feature of few-cycle pulses in contrast to multi-cycle ones is that the temporal evolution of the electromagnetic waveform within the laser pulse is accessible. As virtually all strong field phenomena are directly governed by the electromagnetic field, this feature attracted a great scientific interest in recent years.

The quantity that is used to characterize the evolution of the field within the laser pulse is called the carrier-envelope phase (CEP), which is defined as the offset phase between the peak of the electric field and the peak of the pulse envelope. Stabilization of the CEP of high repetition rate few-cycle sources became possible a few years ago using f-to-2f interferometers, which had an enormous impact on time resolved laser spectroscopy by enabling pump-probe experiments with attosecond resolution. The major limitation of phase stabilization is that it is technically rather complex and it has been demonstrated only up to 0.2 TW peak powers, while few-cycle lasers with multi-10-TW peak powers are already available. These unique laser systems hold promise for extending attosecond metrology and spectroscopy to attosecond control by the generation of energetic isolated attosecond bursts on solid surfaces. Such attosecond pulses would allow for pump-probe experiments with one attosecond pulse releasing the electron and the other controlling its further evolution. However sufficient parameters of the laser pulse's envelope (pulse duration and focused intensity) are already available, due to the lack of phase stabilization conducting waveform dependent experiments at relativistic intensities is still not possible. Therefore the development of a single-shot measurement apparatus that can record the CEP of few-cycle pulses consecutively, and thus would allow for a new measurement method “CEP tagging” with non-phase stabilized lasers, has become a premier challenge in ultrafast optics.

2. Goals

My *first goal* is to perform a complete experimental characterization of a single PM. So far in proof of principle experiments only the time and space integrated (overall) reflectivity was measured which depends on the spatial parameters of the applied laser, therefore such measurements can not be used as an absolute reference. My primary goal was to perform a complete space and time resolved experimental study that can provide the necessary parameters for designing an effective PM system for the 100 TW laser at Laboratoire d'Optique Appliquée.

The experimental characterization aimed for measuring: time and space resolved, time integrated and space resolved (peak) and time and space integrated (overall) reflectivity in function of the incident fluence at various pulse durations. My goal was to measure the plasma triggering threshold at various pulse durations and demonstrate that by applying the optimal fluence on the PM, it can effectively enhance the intensity contrast of high-power laser pulses, while focusability and spatial characteristics of the reflected beam are also improved.

My *second goal* is to set up and fully characterize a double plasma mirror system for the 100 TW laser “Salle jaune” laser at Laboratoire d'Optique Appliquée. My goal was to optimize the fluence on both PMs to exploit the most from the system in order to improve the initial contrast of the laser with several orders of magnitude. My goal was to perform a complete experimental and numerical characterization of the system. In particular I wanted to demonstrate that placing the first plasma mirror into the near field – which is a critical part of the design but unavoidable due to the high pulse energy (2.5 J) – doesn't impair but slightly improves the focusability and more than 40-50% of the laser's initial peak intensity is preserved.

My *third goal* is to develop a single-shot carrier-envelope phase meter. F-to-2f interferometers which are commonly used to stabilize the phase of few-cycle lasers can only detect the rate of change of the CEP, but they are blind to its actual value. Measurement of the CEP therefore requires a completely different approach. In recent years several non-optical methods exploiting various phase sensitive phenomena have been demonstrated to retrieve the CEP, but all of them work only on a proof-of-principle level and require several thousands of laser shots for a single measurement point. Therefore they are applicable only on high repetition rate few-cycle sources with relatively low pulse energies – which are commonplace in many laboratories – as currently only those can be phase stabilized. My goal is to develop the first single-shot measurement apparatus that can record the phase of consecutive non-phase stabilized laser shots, and therefore can be used also on high-power few-cycle lasers, which are non-phase stabilized.

3. Methods of investigation

1. Complete experimental characterization of a single PM was performed in Saclay Laser Interaction Center (SLIC) with the LUCA laser. This required a very complex experimental setup. I've participated in designing and setting up the experiment, participated in the measurements and I performed the data analysis. Laser pulses with 60 fs of pulse duration, 800 nm central wavelength and 100 mJ of pulse energy were focused onto the surface of transparent bulk quartz and anti-reflection (AR) coated quartz targets. To measure the reflectivity of the PM in function of the laser fluence it was varied in a wide range (between 1-100 J/cm^2). Space and time-integrated (overall) reflectivity was measured with energy meters and space resolved time-integrated (peak) reflectivities were obtained by imaging the PM surface on a high-dynamic CCD camera. Time and space resolved reflectivities were measured by chirping the incident pulses to 1.1 ps. The spectrum of the reflected pulse provided the onset of plasma formation at different incident fluences. Distortion of

the beam spatial profile in the far-field and the near-field was measured by imaging the beam in the plane of the PM surface (focal plane), and some distance after the PM.

2. I've participated in the design and construction of the double PM setup, and I completely characterized it. The laser delivers 25 fs pulses with energies up to 2.5 J at 780 nm wavelength with up to 10 Hz repetition rate. Due to the high pulse energy, long focusing mirror had to be used to provide the ideal fluence for the PMs. An f=10 m null telescope created an intermediate focus for the laser beam and recollimated it; near this the PMs were situated both with 45° angle of incidence. The first PM was in the near field 14 cm from the focus and the second PM was in the focus. Due to the improved contrast after the first PM the fluence could had been higher on the second PM. The beam was S polarized and the targets were AR coated quartz. I optimized the fluence by changing the radius of curvature of the deformable mirror. I monitored the beam profiles on the PMs and in the final target plane with webcameras and with a high-dynamic CCD camera respectively. I used an optical beam tracing code to model the DPM setup. I also participated in a short proof-of-principle experiment performed for comparing high-order harmonic generation with and without the DPM system. We observed the generated harmonics with an XUV spectrograph.

3. I've conducted the experiment at Max Planck Institute of Quantum Optics using a few-cycle laser system that delivers sub-4 fs pulses with 800 nm central wavelength at 3 kHz repetition rate. The single-shot stereo-ATI phase meter uses the left/right asymmetry in the yield of high-energy above-threshold-ionization (HATI) electrons along the polarization axis to retrieve the CEP. In order to record the left and right HATI spectra in a single laser a four orders of magnitude increase in the sensitivity was necessary compared to the previous multi-shot apparatus. For that I have made

the following major changes in the new design: I redesigned the magnetic shielding and the vacuum apparatus to increase the number of electrons arriving to the detectors; I changed the data acquisition system from electron counting to digital voltage detection to be able to detect a large number of simultaneously arriving electrons in a single laser shot at high repetition rates; I installed an imaging system that allows for a precise alignment of the laser beam in the interaction volume and optimization of the laser intensity.

4. Results

1. I provided a complete experimental characterization of a single plasma mirror. I measured the peak and overall and the space and time resolved reflectivity of the PM in function of the incident laser fluence at various pulse durations. I measured 74% and ≈60% of peak reflectivity on quartz and AR coated quartz targets respectively at 60 J/cm^2 incident laser fluence. Since the contrast improvement factor is the ratio of the reflectivitiy after and before (0.3% for AR) plasma formation, this provides a contrast improvement factor of nearly 200 [1].

With time resolved measurements I showed that the onset of plasma formation and reflectivity increase happens earlier with increasing fluence, and even at the highest applied fluence it is the rising edge of the main pulse that triggers the PM and not the prepulses or the pedestal. I also demonstrated that the PM acts as a spatial filter and improves the spatial profile of the reflected beam. I measured the plasma triggering threshold and found that it is slightly increasing with increasing pulse duration.

2. I provided a complete experimental and numerical characterization of the double plasma mirror system [2]. I optimized the fluence on both PMs and achieved a 47%-57% of peak and 31% of overall reflectivity for the system. I improved the initial contrast of the 100 TW laser by a factor of $5 \cdot 10^4$ to a record high $5 \cdot 10^{11}$.

I've used an optical propagation code to model the optical transport of the beam and found a good agreement between modelled and measured results including beam profiles on both PMs and focus in the final target plane. Extensively studying the fluence distribution on the first PM, which is in the intermediate field, I found that the beam profile as it was expected is somewhat rough. Although this unavoidably leads to some distortion of the reflected beam profile, this slight degradation is overly compensated by the second PM, which was positioned into the intermediate focus, thus it acts as a spatial filter. Due to this, I observed no degradation of beam quality or focusability in the final target plane, but on the contrary, a slight improvement in the spatial characteristics. As high-power lasers are built to maximize peak intensity on target surface, retaining or rather improving the focusability of the beam is the key feature of the DPM system.

The DPM was engineered as a standard system feature and is easily added or bypassed with a single kinematic stage in the 100 TW Ti:sapphire research laser. The AR targets can be shifted from shot to shot by PC controlled translation stages to provide a fresh undamaged surface for each laser shot.

I've compared high-order harmonic generation with and without the DPM system [4,5]. Temporally cleaning the pulses with the DPM system a narrow beam of harmonics was generated and the laser light was reflected specularly. Without the DPM no harmonics were observed and the reflected beam was diffuse and inhomogeneous. With this I demonstrated that high contrast pulses produced by the DPM system preserves the steep electronic density gradient, which is compulsory for conducting clean laser-solid interactions.

3. I've developed a single-shot stereo-ATI phase meter, which can measure the CEP of few-cycle laser pulses consecutively [3,6]. This I've achieved by increasing the sensitivity with more than four orders of magnitude compared to the previous multishot apparatus. With the single-shot phase meter I recorded the CEP of non-phase stabilized laser pulses consecutively for the first time, which demonstrates that

the phase meter unlike previous mutishot methods doesn't require phase stabilization. This enables a new measurement method CEP-tagging for studying waveform dependent phenomena with any few-cycle laser system, independently whether the particular laser can be phase stabilized or not. This is particularly important as state-of-the-art multi-10TW few-cycle lasers are not phase stabilized. Therefore this new method [7] will potentially open the door for studying the waveform dependence of laser-solid interactions at relativistic intensities.

With the apparatus I recorded the CEP of each shot of a high-repetition rate (3kHz) laser using 40 μJ of pulse energy for the measurement. As it is merely 10% of the pulse energy of a typical multi-kHz few-cycle laser, CEP tagging can be performed also on those systems. Running the laser non-phase stabilized due to the random fluctuation of the phase, CEP tagging will work as an ``ultrafast phase scan'' resulting in an acquisition speed that is orders of magnitude higher than that of regular phase scans. This means that for experiments where the entire 2π phase range have to be studied this novel method is far superior to technically elaborate phase scans.

These results were published in [3] and highlighted in press releases at MPQ [8] and at the TU München [9], and also reported in Laser Focused World [10] and by several popular science websites [11]. The CEP measurement method will be incorporated also into Prof. Rick Trebino's well-known lecture notes on Ultrafast Optics [12].

5. Publications related to the theses

In refereed journals

[1] G. Doumy, F. Quéré, O. Gobert, M. Perdrix, Ph. Martin, P. Audebert, J. C. Gauthier, J. P. Geindre, T. Wittmann,

Complete characterization of a plasma mirror for the production of high-contrast ultraintense laser pulses
Phys. Rev. E **69**, 026402 (2004)

[2] T. Wittmann, J. P. Geindre, P. Audebert, R. S. Marjoribanks, J. P. Rousseau, F. Burgy, D. Douillet, T. Lefrou, K. Ta Phuoc, J. P. Chambaret,
Towards ultrahigh-contrast ultraintnse laser pulses – complete characterization of a double plasma mirror pulse cleaner
Rev. Sci. Instrum. **77**, 083109. (2006)

[3] T. Wittmann, B. Horvath, W. Helml, M. G. Schätzel, X. Gu, A. L. Cavalieri, G. G. Paulus, R. Kienberger,
Single-shot carrier–envelope phase measurement of few-cycle laser pulses
Nature Physics **5**, 357 (2009)

Conference papers

[4] R. S. Marjoribanks, J. P. Geindre, P. Audebert, T. Wittmann, G. Doumy, P. Martin, M. Perdrix, F. Quere, P. Monot, D. Douillet, T. Lefrou, K. T. Phuoc, S. Sebban, J. P. Rousseau, F. Burgy, B. Cross,
Practical Double Plasma-Mirror Pulse Cleaner for High-Intensity Femtosecond Laser-Plasma Harmonics,
2005 CLEO/ QELS refereed conference paper **3**, 2021 (2005)

[5] R. S. Marjoribanks, P. Audebert, J. P. Geindre, T. Wittmann, J. C. Gauthier, Ph. Martin, O. Gobert, M. Perdrix, F. Quere, G. Doumy, P. Monot, P. D'Oliveira, D. Douillet, K. T. Phuoc, S. Sebban, J. P. Rousseau, F. Burgy, B. Cross,
Femtosecond high-intensity laser-plasma harmonics, using practical double-plasma-mirror pulse cleaner American Physical Society,
46th Annual Meeting of the Division of Plasma Physics, 2004APS DPPCO2015M

[6] T. Wittmann, B. Horvath, W. Helml, M. G. Schätzel, X. Gu, A. L. Cavalieri, G. G. Paulus, R. Kienberger,
Single-shot carrier–envelope phase measurement of few-cycle laser pulses
Conference on Ultrafast Phenomena, 2008, THU2P.1

[7] T. Wittmann, M.G.Schätzel, F. Lindner, G.G.Paulus, A Baltuska, M. Lezius, A. Marcinvevicius, F. Tavela, F.Krausz
Carrier-envelope phase measurement of multiterawatt laser pulses
IAMPI, 2006, Szeged, poster P23

Press releases, popular science articles reporting about the single-shot CEP phase meter, which was presented in the thesis:

[8] http://www.mpq.mpg.de/cms/mpq/en/news/press/09_04_19.html

[9] http://portal.mytum.de/pressestelle/pressemitteilungen/news_article.2009-04-18.8174740109/newsarticle_view

[10] Ultrafast-pulse Diagnostics: Single-shot technique measures CEP for few-cycle pulses, Laser Focus World, June 2009

[11] http://www.world-of-photonics.net/link/en/21644171
http://www.laseropto.de/index.php?id=5&artid=3261&np=2&L=1
http://www.analytica-world.com/news/d/99906/
http://idw-online.de/pages/de/news310446
http://www.chemie.de/news/d/99906/

[12] http://www.physics.gatech.edu/frog/lectures/index.html

6. Other publications

In refereed journals

[13] B. Hopp, T. Smausz, T. Wittmann, F. Ignacz,
Comparative time-resolved study of solid-state and liquid ablation of polyethylene-glycol 1000: temperature, viscosity and surface tension dependence
Appl. Phys. B **71**, 315 (2000)

[14] B. Hopp, T. Smausz, E. Tombacz, T. Wittmann, F. Ignacz,
Solid state and liquid ablation of polyethylene-glycol 1000: temperature dependence
Opt. Commun. B **181**, 337 (2000)

[15] Y. Nomura, L. Veisz, K. Schmid, T. Wittmann, J. Wild, F. Krausz,
Time-resolved reflectivity measurements on a plasma mirror with few-cycle laser pulses
New J. Phys. **9**, 9 (2007)

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[16] P. Martin, P. Monot, G. Doumy, S. Dobosz, M. Perdrix, F. Reau, P. D'Oliveira, F. Quéré, P. Audebert, J. P. Geindre, J. C. Gauthier, T. Wittmann,

Utilization of a plasma mirror for the production of high-order harmonics from a planar surface
Laser Optics 2003: Superintense Light Fields and Ultrafast Processes; Proceedings of the SPIE **5482**, 55. (2004)

[17] L. Veisz, Y. Nomura, K. Schmid, F. Krausz, T.Wittmann,
Plasma mirror with few-cycle laser pulses
2005. CLEO/Europe page:422 (2005)

[18] F. Tavella, T. Wittmann, K. Schmid, B. Horvath, A. Cavalieri, L. Veisz, A. Marcinkevicius, F. Krausz,
Stronger seed for a multiterawatt few-cycle pulse OPCPA
2007. CLEO/Europe page:422 (2005)