Terawatt-intensity few-cycle laser pulses

Witte, S.M.

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Summary

An important topic in laser physics is the ability to control and manipulate the electric field of light, and to generate ever shorter light pulses with a controlled waveform. Such pulses allow the construction of an "ultra-high-speed camera" that can follow the fastest processes in nature. In principle, the shortest pulse that can be produced will contain at least one half-cycle of the electric field. This implies that faster pulses can only be made with light at a shorter wavelength, such as extreme-ultraviolet or soft-X-ray radiation. The production of ultrashort X-ray pulses can be achieved through high-harmonic generation in a gas, using powerful laser pulses with a controlled electric field.

Another challenge in atomic physics is the measurement of absolute transition frequencies in atoms, ions and molecules with ever increasing precision. Such high-precision measurements allow a direct comparison between experimental observations and theoretical predictions about the structure of matter. In addition, they can lead to more accurate atomic clocks, and could be used to detect possible changes of fundamental constants over time. While the invention of the frequency comb has revolutionized precision measurements, its impact has remained limited to the ultraviolet, visible and near-infrared spectral regions where narrow-band continuous-wave lasers are available. One of the goals of this thesis is to develop methods for performing frequency comb spectroscopy at much higher frequencies. This frequency range (known as the extreme-ultraviolet) contains many atomic and ionic transitions that are of fundamental physical interest, but narrow-band continuous-wave lasers are not available at such high frequencies.

To achieve these goals, we have constructed a laser source that can produce ultrashort laser pulses with extreme peak intensity, while maintaining the phase coherence and the frequency comb spectrum of these pulses. Phase-locked pairs of these powerful broadband pulses can then be used for high-resolution frequency measurements. In addition, these intense ultrashort phase-controlled laser pulses are well-suited for the production of coherent ultrashort soft-X-ray pulses with a duration below 1 femtosecond (10^{-15} seconds). A more detailed background and motivation for the work that is presented in this thesis is given in chapter 1, along with an outline of its contents.

Besides providing a theoretical background on the physics and propagation of ultrashort light pulses, chapter 2 introduces all aspects of the frequency comb laser system that we have constructed. This frequency comb forms the basis of our spectroscopy experiments, which will be discussed in chapter 3. The modes of the frequency comb have been stabilized to a GPS-disciplined Rb-clock, allowing a frequency accuracy of 10^{-11} in a 10 second averaging time. The carrier-envelope phase shift between pulses has been stabilized to better than 1/40$^{\text{th}}$ (RMS) of an optical cycle using f-to-$2f$ interferometry and fast electronics.

In chapter 3, a proof-of-principle experiment is presented that demonstrates the feasibility of high-resolution frequency metrology with broadband laser pulses. We

show that pairs of phase-locked pulses from our frequency comb oscillator can be amplified and frequency-upconverted without losing pulse-to-pulse phase coherence, and that these pulses can be used for accurate frequency measurements. We exploit the phase coherence between the individual laser pulses to induce quantum interference in krypton atoms, coherently exciting one particular atomic transition under study. From a measurement of the final excited state population as a function of the phase and time delay between the laser pulses, the transition frequency can be deduced. We have performed an absolute frequency calibration of the krypton $4p^6 \rightarrow 4p^5 5p[1/2]_0$ two-photon transition in the deep-ultraviolet spectral range, at a wavelength of 2×212.55 nm. An absolute frequency accuracy of 3.5 MHz (1×10^{-9} relative accuracy) has been achieved, using powerful picosecond laser pulses that have a spectral bandwidth of about 1 THz and a peak intensity of several megawatts. Isotope shifts have been measured with 150 kHz accuracy. This method of measuring transition frequencies combines the high peak intensity of the individual pulses with the high resolution given by the total duration and pulse-to-pulse phase coherence of the full pulse train. Therefore, it enables high-resolution frequency comb spectroscopy in wavelength ranges where narrow-band continuous-wave lasers are not available.

Now that this method of performing frequency metrology has been proven successful, an important next step to advance this technique further is the construction of suitable laser systems that can provide intense and phase-stable laser pulses. A promising candidate for the amplification of ultrashort laser pulses to extreme intensity without the loss of phase coherence is noncollinear optical parametric chirped pulse amplification (NOPCPA). This technique is based on the process of optical parametric amplification (OPA), in which a high energy photon is split into two photons of lower energy inside a nonlinear optical medium. This OPA process can effectively work as an amplifier by seeding the nonlinear interaction with a weak laser pulse containing photons with the right energy. Energy transfer can then take place from a high-power pump pulse to this low-power seed pulse. Optical parametric amplification can be scaled up to produce extremely high-intensity pulses by combining it with the principle of chirped pulse amplification (CPA). In a CPA scheme, an ultrashort pulse is first stretched in time by a dispersive delay line, known as a pulse stretcher, to lower the peak intensity before amplification. This stretched pulse is amplified and subsequently recompressed in a second delay line with dispersion of opposite sign compared to the stretcher. The combination of OPA and CPA in a noncollinear geometry is known as NOPCPA, and this technique has several interesting properties that should allow the phase-stable amplification of extremely broadband laser pulses to high intensity.

The theory of how OPA works is given in the first part of chapter 4. For the second part of this chapter, simulations on a realistic NOPCPA amplifier system have been performed, to find optimal working values for many important parameters in such a system. We have developed a computer simulation program based on the split-step Fourier algorithm, which incorporates both dispersion and nonlinearity during pulse propagation through multiple amplification stages. A good agreement with the experimental situation described in later chapters is found. We find various effects that are specific to NOPCPA (i.e. that do not occur in conventional population-inversion-based laser amplifiers), such as wavelength-dependent gain saturation and the need for tight synchronization of pump and seed pulses.

To develop the desired amplifier system for few-cycle terawatt laser pulses, several different NOPCPA systems have been constructed and systematically investigated. To allow such an investigation, we constructed various auxiliary optical systems, such as a stretching and compression system for few-cycle laser pulses with a spectral throughput of 400 nm, and an Nd:YAG-based pump laser source that delivers up to 160 mJ per pulse at a wavelength of 532 nm in 60 ps pulses. In addition, the required diagnostic tools have been designed and constructed, such as a SPIDER system to measure the pulse duration with sub-femtosecond resolution, as well as setups for the characterization of pulse contrast and phase stability. Most of these parts of the total NOPCPA system are described in detail in chapter 5.

The three different experimentally realized NOPCPA implementations are then discussed in chapters 6–8. In chapter 6, an NOPCPA system is presented that produces 0.1 mJ, 11.8 fs pulses at 1 kHz repetition rate. Using Fourier-transform spectral interferometry, quantitative measurements on the phase stability of the amplified pulses have been performed. We find that the NOPCPA amplification adds less than 100 mrad phase noise ($<1/60^{th}$ of an optical cycle) to our pulses, confirming that NOPCPA can indeed be used for the amplification of phase-locked frequency comb pulses.

Chapter 7 presents a system that takes the pulse intensity to a much higher level: Using the NOPCPA technique, we have constructed the first laser system that produces sub-10 fs pulses with an intensity exceeding a terawatt, at a repetition rate of 30 Hz. The total fluorescence is kept well below 1%. Since only three amplification stages are required to reach terawatt intensity, and the required stretching ratio of the pulses is only $\sim 10^3$, the amplifier system can be kept remarkably compact, and currently stands as one of the smallest terawatt amplifiers in the world.

The proof-of-principle terawatt system from chapter 7 has been upgraded to provide even shorter pulses at higher intensity, as explained in chapter 8. By seeding the amplifier with a new, home-built ultrabroadband Ti:Sapphire oscillator capable of producing 6.2 fs pulses, the amplifier output could be pushed to its theoretically predicted limit in terms of spectral bandwidth. In addition, an improved pump laser design is used to boost the power further. This system generates 7.6 fs pulses with an intensity of 2 TW. These pulses are compressed to within 5% of its Fourier-limited pulse duration of 7.3 fs, using grating-based stretching and recompression combined with adaptive spectral phase shaping. With an OPA-based pulse contrast measurement setup suited for few-cycle laser pulses, we found a pre-pulse contrast of 2×10^{-8} between the main pulse and the fluorescence background.

Chapter 9 concludes this thesis. In this chapter, several options are presented to optimize the pump pulse shape for NOPCPA, based on time-multiplexing and pulse-tilting. A new parametric amplification scheme is proposed, which we have named optical parametric tilted pulse amplification. This implementation of parametric amplification may provide a compact and flexible setup for frequency comb spectroscopy with ultrashort laser pulses. Finally, the current status of frequency comb spectroscopy in the extreme-ultraviolet is reviewed, and the prospects of precision measurements using our NOPCPA system are discussed.