

# Ultrafast double pulse parametric amplification for precision Ramsey metrology

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**Abstract.** An optical parametric chirped pulse amplifier system for pulse pairs is presented. The differential phase stability of the pulse pairs is 20 mrad, giving good prospects for high resolution Ramsey spectroscopy in the extreme ultraviolet.

## Introduction

The convergence of ultrafast optics and high precision optical frequency metrology has led to fascinating new possibilities. The advent of femtosecond laser optical frequency combs (OFC) [1] turned the determination of arbitrary optical frequencies into a routine task. This enabled new high accuracy test of quantum electrodynamics (QED) [2] and the determination of tight lower bounds on the current time dependence of fundamental physical constants [3] as well as it opened the possibility of creating optical frequency standards which have the potential to give 18 digits of accuracy. Simultaneously OFC opened the door to attosecond physics [4].

In recent years a new branch of high-precision physics is emerging from the crossfertilization between laser frequency metrology and ultrafast technology. Ultrashort pulses provide an extremely high peak power suitable for relatively efficient nonlinear frequency conversion of the available power into frequency ranges where no laser sources exist. If these pulses originate from a phase stable OFC, they interfere to create a frequency comb inside the THz bandwidth of a single femtosecond pulse. Each of the modes of this frequency comb can be extremely narrow and are suitable as a probe for high resolution spectroscopy experiments. As the nonlinear conversion can preserve this comb structure, high-precision experiments in the extreme ultraviolet spectral range come into reach, and first proof of principle experiments have been performed to demonstrate this idea [5,6].

Quite a few interesting atomic transitions exist in the extreme ultraviolet (XUV) wavelength range below 100 nm. As an example, the  $1s^2\ ^1S_0 - 1s4p\ ^1P_1$  transition in atomic helium at 52 nm wavelength could be used to improve the value of the ground state Lamb shift in this atom by at least an order of magnitude. One specifically intriguing possibility is to determine the frequency of the 1s-2s transition in He II, a 2-photon transition at 60 nm. He II is a Hydrogen like system which is specifically simple to model in QED, and comparisons between theory and experiment could in principle be carried out at an extreme level of accuracy. Compared to Hydrogen, the ground state energy in He II is four-fold lower which leads to stronger relativistic effects. Specifically, the ground state Lamb shift (i.e. the QED correction to the Dirac energy level structure) is 16 fold stronger than in Hydrogen.

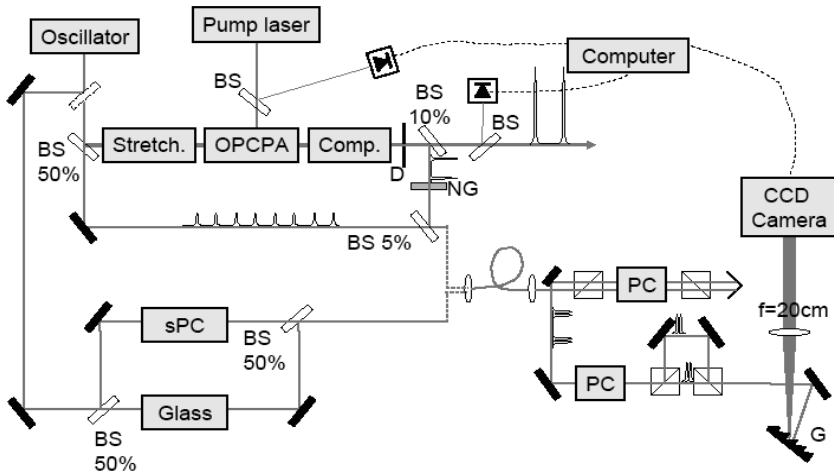
Currently, there exist two approaches to coherent XUV generation for high resolution spectroscopy. The first possibility is to enhance the entire bandwidth of a MHz repetition rate femtosecond frequency comb laser in a passive optical resonator

in order to achieve the intensities required for high order harmonic generation (HHG) [7]. On the other hand it can be shown that optical Ramsey type spectroscopy, where only two identical pulses with a variable but precisely known delay and carrier envelope phase are used to excite the atom, is essentially equivalent. Such pulse pairs can be obtained by pulsed amplification of an OFC seed source. While the former is a continuous wave technique which avoids many systematic effects due to transients in the optically active materials in the setup, we prefer the latter as it can rely on cutting edge non collinear optical parametric chirped pulse amplifier (NOPCPA) technology [8]. This offers more than three orders of magnitude larger peak power, so that the nonlinear conversion becomes a lot more efficient, facilitating the actual spectroscopy experiment.

### Phase-stable double pulse NOPCPA

The double-pulse NOPCPA is based on the single-pulse system presented previously [9]. To achieve double-pulse amplification, the pump pulses are split, delayed with respect to each other and superimposed in a symmetrized relay imaged Mach-Zehnder

interferometer. In this way two identical (in spatial profile) pump pulses with a delay of 6.6 ns are generated. This delay matches the time between two pulses from our 151 MHz OFC seed oscillator. In this way we can amplify a pair of subsequent pulses from our OFC to the millijoule level with a pulse duration down to 10 fs. This pair can be upconverted into the XUV spectral and used for a Ramsey experiment with a spacing of the Ramsey fringes of 151 MHz.



**Fig. 1.** NOPCPA and Mach-Zehnder interferometer for measurement of the differential phase shift accumulated during the amplification in a NOPCPA, and the setup used to test the reliability of the measurement (dotted beam line). BS: Beam splitter, PC: Pockels cell, sPC – slow Pockels cell, NG: neutral grey filter, Comp.: compressor, G: 1200 l/mm grating

If the wavefront and the intensity of the two pulses is not identical, the two amplified pulses can acquire different phase shifts during the amplification process. This would lead to a shift of the Ramsey fringes with respect to the original comb mode positions. Such a shift is multiplied up by the upconversion process, which means that it needs to be accurately measured and monitored.

Figure 1 shows a schematic of the setup used to monitor differential phase shift induced by the NOPCPA system on the amplified pulse pair. It consists of a Mach-Zehnder interferometer including the NOPCPA, the stretcher and compressor in one arm. The output of this interferometer is analysed using spectral interferometry. In order to maintain a good contrast in the interference signal, the output of the interferometer is spatially filtered using a single mode optical fiber and temporally gated to remove the pulses from the seed oscillator which have no amplified counterpart using a double passed Pockels cell (PC1). Like this we obtain a fringe contrast of almost unity with a background from leaking oscillator pulses of less than  $10^{-4}$ . The two remaining pulses are spatially split using a second Pockels cell so that two interferograms can be recorded, one for each pulse. The information on the differential phase shift is now contained in the relative position of the two spectral interferograms. As only differential phase shifts are of importance for the spectroscopy the absolute position of the two interferograms is not important and the interferometer needs only to be stable on a 10 ns time scale (the separation between the pulses). This is passively guaranteed even for the almost 10 meter arm length of the interferometer.

## Results

The accuracy of the phase measurement scheme was found to be better than 10 mrad. The single shot stability of the interferometer output is on the same order of magnitude, so that shot to shot fluctuations can be accurately measured and taken into account. The rms phase stability of the NOPCPA system was found to be 20 mrad. This is already sufficient for the observation of clear Ramsey fringes even at the 15<sup>th</sup> harmonic of the fundamental signal frequency at 800 nm, that is required for the  $1s^2\ ^1S_0 - 1s4p\ ^1P_1$  transition in He I. The achievable accuracy for the Lamb shift due to the amplifier phase shift is therefore at least 15 MHz which would be a threefold improvement over previous results.

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