Frequency comb metrology at PHz frequencies: precision in the extreme ultraviolet

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Abstract

The capability of frequency-comb (FC) lasers to precisely measure optical frequencies is extended to the multiple-PHz domain. This frequency region, which covers the extreme ultraviolet (XUV, wavelengths shorter than 100 nm), was previously not accessible to these devices. Frequency comb generation is shown for 51-85 nm by amplification and coherent up-conversion of a pair of pulses originating from a near-infrared femtosecond FC laser. Moreover, Ramsey-like signals with up to 61% contrast are observed when the XUV comb is scanned over transitions in argon, neon and helium, resulting in an 8-fold improved determination of the ground state ionization energy of helium.

1. Introduction

Frequency comb (FC) lasers [1,2] have greatly advanced fields such as precision spectroscopy, optical atomic clocks, and attosecond science. FC lasers provide a phase-coherent link between optical frequencies of several hundred THz, which cannot be measured directly, and radio frequencies that can readily be counted and compared to frequency standards.

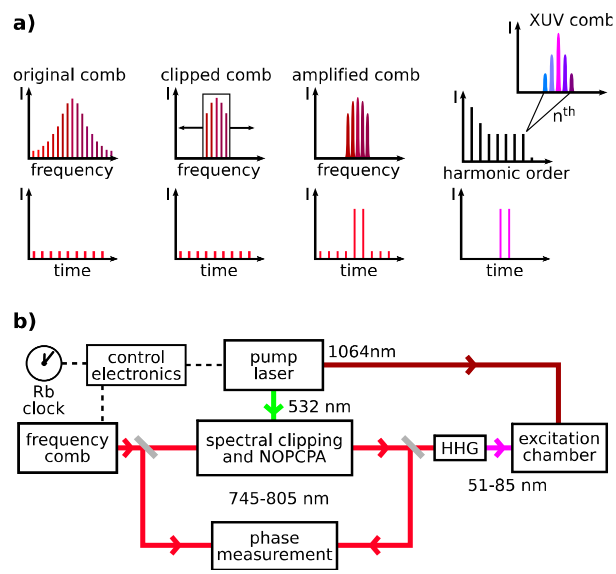

Fig 1. (a) Spectral and temporal structure of the different stages of XUV comb generation and spectroscopy (left to right): continuous coherent pulse train from the FC laser, the clipped FC spectrum in the stretcher, the cosine-modulated spectrum of a pair of amplified pulses, and odd harmonics of the amplified FC laser pulses, each containing a cosine-modulated XUV comb corresponding to the XUV pulse pair. (b) Schematic of the experimental setup; NOPCPA: non-collinear parametric chirped pulse amplifier, HHG: high-harmonic generation.

A FC effectively acts as a gearbox between these two frequency domains, so that optical frequencies can be counted electronically. This is possible because all the emitted modes (easily up to a million) of a comb laser are equidistant in frequency, and can be described by only two numbers, f_{CEO} (the carrier-envelope offset frequency) and the repetition frequency f_{rep}, both in the RF domain. In the frequency domain a comb-like spectrum is seen, where the m^{th} mode has a frequency equal to:

$$f_m = f_{CEO} + m\, f_{rep} \tag{1}$$

Viewed in the time domain, the laser emits an infinite train of pulses at the repetition rate f_{rep}, with a phase slip between subsequent pulses (as measured between the optical carrier and the pulse envelope) of $\Delta\phi = 2\pi f_{CEO}/f_{rep}$.

We demonstrate that the remarkable precision and structure of frequency combs can be transferred to extreme ultraviolet (XUV) wavelengths by parametric amplification and high-harmonic generation (HHG) of two subsequent comb laser pulses (see Fig. 1b). In effect a pair of phase-locked extreme ultraviolet pulses is generated, which can be used directly for precision spectroscopy without the need for an additional spectroscopy laser. Viewed in the frequency domain, the spectrum of the upconverted pulse sequence in the XUV resembles again a frequency comb, but now in the form of a cosine-modulated spectrum (see Fig. 1a). From a time-domain perspective, excitation with phase-locked pulses is a form of Ramsey excitation (see e.g. [3,4]).

2. Experimental methods

The setup (Fig. 1b) consists of a Ti:Sapphire FC laser which is referenced to a GPS-controlled rubidium frequency standard. From this source of phase controlled pulses, a center wavelength and bandwidth of phase-controlled pulses is selected with a slit placed in the stretcher that is part of the parametric amplifier system. The resulting 'clipped' comb laser pulses can be tuned from 745 nm to 805 nm. A dedicated non-collinear chirped pulse parametric amplifier (NOPCPA) is used to amplify two subsequent (and spectrally clipped) pulses from the comb laser to 5 mJ per pulse. After compression 1-2 mJ per pulse is available for HHG. The phase distortion by the amplification process (typically on the order of 100 mrad) is measured by spectral interferometry and is taken into account [5]. The XUV comb is then produced by HHG of the two pulses in a noble gas jet. By selecting different harmonics (the 9^{th} to 13^{th} harmonic made in xenon, the 15^{th} made in krypton) and the center wavelength from the FC, XUV comb generation is made possible over a range of 85 nm down to 51 nm. The bandwidth in the IR of the comb pulses is set between 3 and 6 nm to ensure that the harmonics produced after amplification and HHG are narrow enough to excite only one transition at the time.

The coherence of the generated XUV light has been verified by exciting one-photon transitions in argon, neon and helium. For this purpose the XUV beam is crossed perpendicularly with a low divergence (< 4 mrad) atomic beam.

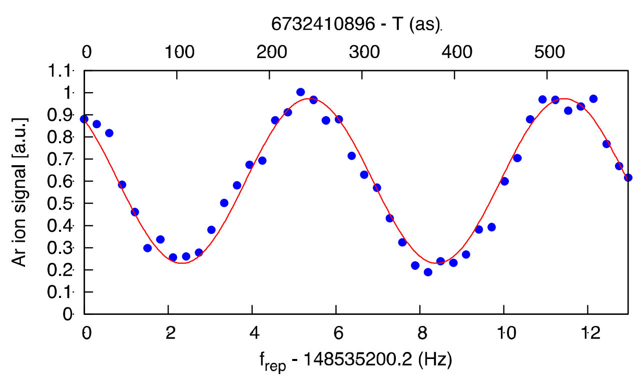

Fig 2. Measured excitation probability (circles) of argon at 82.53 nm from the ground state to $3s^2\ 3p^5(^2P_{1/2})6s[1/2]_1$ excited state, normalized by the XUV pulse energy, as a function of the repetition rate f_{rep} of the frequency comb laser. A pure argon beam is used, and the red line represents a sinusoidal fit to the data. The upper scale shows the time delay between the pulses in attoseconds.

After excitation with the two XUV pulses, the excited state atoms are ionized with an infrared laser pulse at 1064 nm and detected in a time-of-flight spectrometer. By changing the delay between the XUV pulses (i.e. the repetition rate of the frequency comb laser) in approximately one-attosecond steps, the cosine-shaped comb spectrum is scanned over the transition. The recorded signals are binned in 20 equidistant groups over a Ramsey period, which is equal to the mode spacing, and therefore the repetition rate of the frequency comb laser. In Fig. 2 a typical resulting signal is shown for argon excited at 82.53 nm from the ground state to the $3s^2\ 3p^5(^2P_{1/2})6s[1/2]_1$ excited state. The XUV comb was generated in this case by the 9th harmonic in xenon. The argon signal shows a high contrast of 61% for a repetition rate of the FC of 148 MHz. Similar spectra have been recorded for neon near 60 nm (13th harmonic) and for helium (15th harmonic in krypton) near 51 nm.

3. Results

In the case of helium, recordings were made of 4 different transitions ($1s^2\ ^1S_0$ - $1snp\ ^1P_1$ with n = 4-7). The signals for n=4,5 were used to obtain an improved value for the ground state ionization energy, by measuring the ion signal together with the parameters of the frequency comb laser, the phase shift between the amplified pulses, and the energy of the IR and XUV pulses. Fig. 3 shows a typical He ion signal as a function of pulse delay for f_{rep}=185 MHz. From the fit of the recorded signals precise transition frequencies can be deduced. Because the spectroscopy signal is periodic with f_{rep} the measured transition frequency can only be determined up to an integer multiple of the repetition frequency. To resolve this ambiguity, the measurements in helium were repeated for different repetition rates of the laser between 100 MHz and 185 MHz. The correct 'comb mode' is located where the possible transition energies for all repetition frequencies coincide. Furthermore, a careful analysis of systematic effects was performed. Differential spatial and temporal pulse deformation of the amplified IR double pulse were monitored and controlled using the aforementioned spectral interferometric measurement technique [5], with an accuracy better than a 200th of an optical cycle. Many other systematic effects have been investigated as well, such as effects of ionization and adiabatic phase shifts during the HHG process, self-phase modulation in optics, and Doppler shifts. Using the precisely known 4p and 5p excited state energies, we find the ground state energy (ionization potential) of helium at 5945204212(6) MHz [6]. This is in good agreement with the most recent theoretical calculations [7,8]. The obtained accuracy is nearly an order of magnitude better than previous experiments where the ionization potential of helium was determined using single nanosecond-duration laser pulses [9,10], which clearly demonstrates the power of XUV frequency comb excitation.

The contrast of the signals in helium have been analyzed using a model that includes the effects of the lifetime of the excited state, Doppler broadening, XUV pulse energy ratio, direct ionization background, and the time delay between the pulses. From this analysis it is found that the XUV comb at 51 nm has a 50 attosecond rms excess timing jitter which can be attributed to the HHG process. A possible cause of this jitter is intensity fluctuations of the IR pulses (~ 5%), and fluctuations in density and level of ionization in the HHG gas jet.

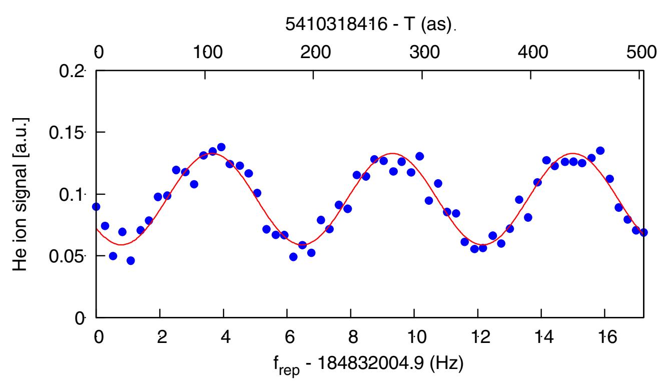

Fig 3. Measured excitation probability (circles) of helium at 51.5 nm on the $1s^2\ ^1S_0 – 1s5p\,^1P_1$ transition, normalized by the XUV pulse energy, as a function of the repetition rate f_{rep} of the frequency comb laser. In this example f_{CEO} is locked at 46.21 MHz, and a 1:5 He:Ne mixture is used for the atomic beam. The red line represents a sinusoidal a fit to the data. The upper scale shows the time delay between the pulses in attoseconds.

4. Conclusions

XUV frequency comb generation has been demonstrated from 85 nm to 51 nm based on amplification and upconversion of two FC laser pulses. With this comb, the first absolute frequency measurement has been performed in the XUV, resulting in an 8-fold improved helium ground state ionization energy. Improvements to the laser setup are in progress to make the frequency comb amplification and HHG process better controlled. This should reduce the measured timing jitter of 50 attoseconds between the two upconverted comb pulses, so that comb generation could become possible at even shorter wavelengths such as soft X-rays.

5. Acknowledgments

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