Demonstration of Frequency Comb Laser Spectroscopy in the Vacuum-Ultraviolet

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Abstract. High-resolution spectroscopy at 125 nm is performed on xenon, using amplified frequency comb pulse trains that are frequency up-converted in a gas cell. It is shown that the phase coherence in the up-conversion process is maintained to better than $1/30^{th}$ of a VUV cycle, thereby demonstrating the potential for sub-MHz accuracy in the vacuum-ultraviolet.

1. Introduction

An alternative technique for precision spectroscopy that is rapidly gaining interest is the direct excitation of atomic transitions with a train of pulses from a frequency comb laser. As the high peak intensity of such ultrashort pulses allows efficient up-conversion of the infrared frequency comb output to higher frequencies, this technique seems very promising for extending high-resolution spectroscopy to the VUV and XUV. The feasibility of frequency combs in the XUV has been demonstrated by employing external enhancement cavities to increase peak intensity, with intra-cavity harmonic generation using a gas jet [1,2]. As amplified pulses can in principle generate shorter wavelengths and can have higher photon yield per pulse, we investigate Ramsey-type quantum interference spectroscopy with a train of amplified and up-converted frequency comb pulses ([3] and references therein).

2. Experimental Setup and Results

A train of 2 to 6 pulses from a frequency comb oscillator is selected, which are boosted to tens of microjoules energy per pulse in a multipass Ti:Sapphire amplifier. We generate VUV radiation at a wavelength of 125 nm by second harmonic generation of the 750 nm amplifier output in a BBO crystal, and subsequent third harmonic generation (THG) in a gas cell filled with either oxygen or acetylene (see Fig. 1). The produced VUV pulse train perpendicularly intersects an atomic beam of xenon to excite the $5p^6\ ^1S_0 \rightarrow 5p^5(^2P_{3/2})5d[1/2]_1$ transition. The final excited state population is probed by ionization with a delayed 532 nm laser pulse, and detection of the produced ions with a time-of-flight mass spectrometer. The quantum interference in the excitation process is

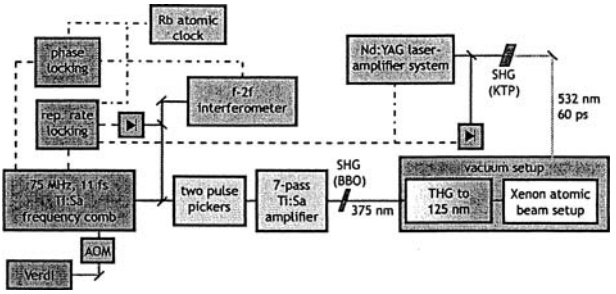

Fig. 1. Setup for 125 nm quantum interference spectroscopy on xenon

made visible by scanning the delay between pulses while keeping the phase difference between pulses fixed (Fig. 2). The initial time delay is 13.3652 ns, leading to a fringe period of 74.8212 MHz, as is shown in the first panel of Fig. 2a. This time delay is changed by 12.6 attoseconds per scan step, and every data point is averaged over 10 seconds (10,000 shots). By adding more pulses to the first two, the frequency resolution can be increased, as is shown in Fig 2a: in the limit of adding an infinite number of pulses and infinite transition lifetime, sharp modes will emerge, resembling the original frequency comb spectrum. We have performed measurements with pulse trains of up to six pulses. The fringes retain the 74.8 MHz period, and narrow down as expected when more pulses are added.

However, the amplifier induces small phase deviations for longer pulse trains, leading to small shifts of the fringe position and asymmetries in the fringe shape. Such phase shifts are difficult to measure for longer pulse trains. When using only two pulses, measurements of amplifier-induced phase shifts can be performed with an accuracy better than 25 mrad, as we have shown previously [3]. In addition, any phase shift caused by THG will simply show up as a shift of the two-pulse interference pattern instead of causing a complicated fringe shape. The simplicity of two-pulse measurements can be combined with the high frequency resolution of longer pulse trains by selecting pulse pairs with a larger time delay from the oscillator output. An initial measurement with small time delay can then be used to identify the comb mode through a comparison with existing low-resolution spectroscopic data, after which subsequent measurements with increasing pulse separation can be performed to increase the resolution by zooming in on the transition. Results of such an experiment are shown in Fig. 2b. The narrowest resonance that is observed has a full width at half maximum (FWHM) of 7.5 MHz, which constitutes an order of magnitude improvement compared to any other measurement performed so far at such short wavelengths by using nanosecond pulsed lasers [4] or cw laser sources [5]. The fringe position can be determined with 40 kHz accuracy, and a total uncertainty of 270 kHz is obtained when phase shifts caused by the amplifier are taken into account, translating into a measurement accuracy of 1×10^{-10} (excluding Doppler and AC Stark effects). The absolute transition frequency has not been calibrated, as no sufficiently accurate previous measurement is available to identify the proper

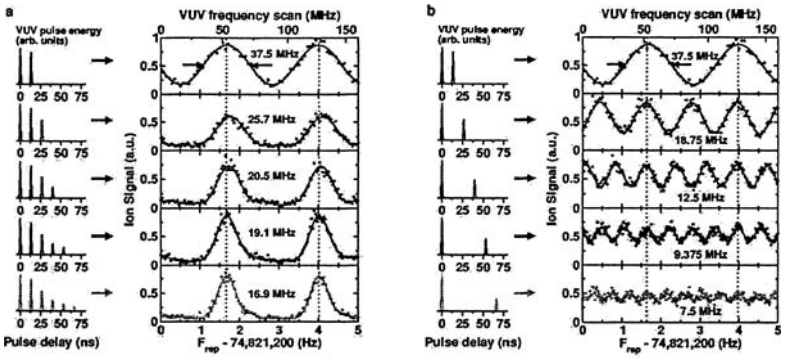

Fig. 2. Quantum interference signals at 125 nm in ^{132}Xe. **(a)** Measured signals using a VUV pulse train of increasing length. **(b)** Measured signals using two VUV pulses with increasing delays. The width of the resonances (FWHM) is denoted in each panel.

mode at present, in contrast to our previous experiment on krypton [3]. To check for possible phase shifts due to the harmonic generation process, two-pulse signals where compared for different THG gas densities and harmonic yield. For both for oxygen and acetylene as the THG medium, no phase shift is observed within the measurement accuracy of 1/30th of a VUV optical cycle.

3 Conclusions

The results [6] confirm the exciting prospects of frequency comb metrology in the VUV and XUV, and are a significant step forward towards our goal to perform sub-MHz quantum interference metrology on the $1s^2$›1s2s two-photon transition at 120 nm in helium, and similar transitions in e.g. He^+ to measure QED and nuclear size effects directly from the ground state.

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