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Towards high-precision VUV frequency comb spectroscopy of the thorium-229 nuclear isomer

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Abstract

The radio isotope thorium-229 is predicted to possess a unique, extremely low-energy metastable excited state of the nucleus in the range of 7.8 ± 0.5 eV, offering the chance to coherently manipulate a nucleus by (VUV) laser light for the first time. Apart from exciting fundamental research questions (including the search for temporal variations of fundamental constants), this well-shielded narrow-linewidth transition opens up the possibility to realize a compact solid-state optical time standard, surpassing existing systems by orders of magnitude in both precision and accuracy.

Since optical (THz) frequencies can't be directly measured using electronic equipment, an optical "clockwork" is needed to exactly divide them down to the radio frequency regime, where individual oscillations can be counted and compared to existing standards (like cesium atomic clocks). This problem arises for all types of optical clocks and has been recently solved by the advent of optical frequency combs. These specially stabilized femtosecond lasers provide a broad tunable spectrum of exactly equidistant narrow-linewidth frequency components, which can be thought of as a "ruler in frequency space", usable as a reference for precise beat-note measurements of any unknown optical frequency. In order to apply these versatile tools to a potential "thorium clock", existing femtosecond laser sources - which operate in the visible to near-infrared domain - somehow have to be converted to the 159 ± 10 nm range where the nuclear transition is expected.

Our approach to this frequency-upconversion (and scope of this thesis) is the generation of the 5th harmonic of a titanium-sapphire laser based comb, with a central wavelength of 800 nm, by means of high harmonic generation (HHG) in a xenon gas jet, inside a passive external enhancement resonator. This allows to maintain the short pulse length of around 25 fs and the high repetition rate (108 MHz) of the source. It has also been already demonstrated that the phase coherence and thereby the comb structure of the source is conserved in the process.

However, although conceptually simple, the setup involves several fundamental challenges: The highly nonlinear process of HHG has a very low conversion efficiency, and the required high peak power density above 10^{13} W/cm² is barely reached inside the enhancement cavity on the basis of only 900 mW of available fundamental power, even with a comparably large power enhancement factor of 250. As already implied by the name vacuum ultraviolet (VUV), light at the target wavelength is absorbed by oxygen in air, so that everything has to be operated under vacuum conditions and remote-controlled. Both carrier-envelope offset frequency and pulse repetition rate have to be stabilized to the cavity. Chromatic dispersion effects, leading to pulse chirp and temporal broadening, have to be strictly controlled. Generally, (high-finesse) femtosecond enhancement cavities also have very strict requirements on mechanical stability. Efficient outcoupling of the (collinearly) generated harmonic light from the cavity is a non-trivial task, as is the optimization of phase matching and the gas jet geometry.

To ensure a tight intracavity focus and clean Gaussian beam profiles of the generated harmonics, we introduce a novel three-dimensionally folded variant of the typical planar bow-tie resonator geometry. This scheme allows us to mitigate the formation of an elliptical intracavity focus, usually caused by oblique incidence on spherical focusing mirrors, while still maintaining linear polarization. Third and fifth harmonic signals are successfully generated with average powers of 20 μ W and 60 nW respectively.