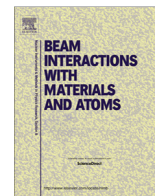




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## Towards precision measurements on highly charged ions using a high harmonic generation frequency comb

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## ABSTRACT

Highly charged ions (HCI) offer many advantages over neutral and singly charged ions for probing fundamental physics. Recently they have been proposed as candidates for novel frequency standards. The project presented here aims at studying HCI with high precision in the extreme ultraviolet (XUV) region, where many of their transitions are located. To this end, an XUV light source is being developed, using a stabilized frequency comb to generate high-order harmonics inside the focus of an enhancement cavity. This optical resonator resides in an ultra-high vacuum (UHV) chamber and is designed to have a very tight focus. The generated XUV light will be guided to a cryogenic linear Paul trap, where trapped HCI are sympathetically cooled by Be<sup>+</sup> ions. Individual comb lines can then be used to drive narrow transitions in HCI, enabling XUV spectroscopy with unprecedented accuracy.

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## 1. Introduction: HCI as probes for fundamental physics

Recent theoretical works have raised the interest in highly charged ions (HCI) by suggesting their use for future frequency standards [1–4]. The current generation of atomic clocks reaches fractional accuracies of 10<sup>−18</sup> [5], limited mainly by influences of external fields which induce shifts in the transition frequencies of the trapped atoms or ions. In HCI, the spatial extent of the electron wavefunctions is much smaller and their sensitivity to external perturbations is therefore significantly decreased. This opens possibilities for reaching relative accuracies in the range of 10<sup>−19</sup> [2]. Furthermore, specific HCI have been predicted to possess an extremely high sensitivity to possible variation of the fine-structure constant [6].

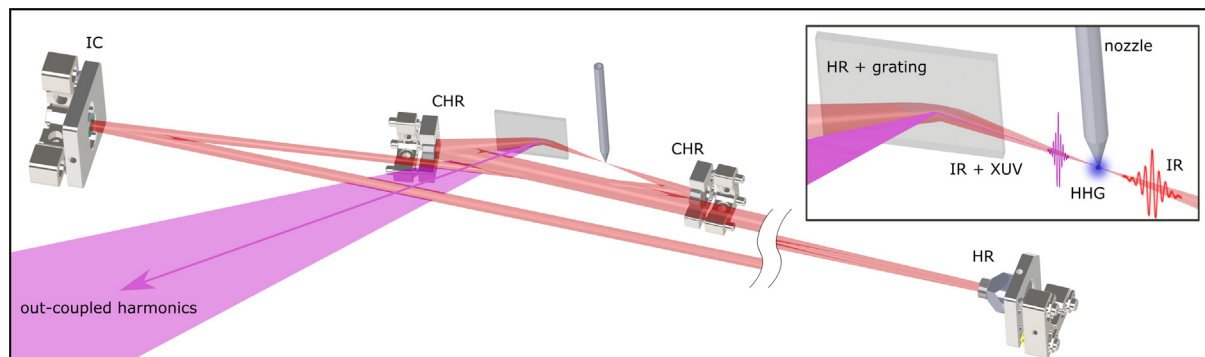
In order to exploit the advantages of HCI, one needs to determine their transition energies with the highest possible accuracy. However, precise theoretical calculations on these energies are complex, due to limitations of perturbative approaches. Currently, the highest theoretical accuracy for a multi-electron system, roughly 0.01%, is reached in the case of boron-like ions [7,8], while for more complex systems like Ir<sup>17+</sup>, they are only at the 1% level [6]. For those reasons, experimental searches are mandatory.

HCI can be produced in an electron beam ion trap (EBIT), where a dense electron beam, compressed and guided by a magnetic field, ionizes and traps the ions. Due to the constant flux of high-energy electrons, temperatures in the order of 10<sup>6</sup> K prevail in the trap, restricting the accuracy in frequency determinations to ppm levels [9]. The cryogenic Paul trap experiment (CryPTEx) at the MPIK has recently succeeded to overcome these limits by extracting HCI from an EBIT and subsequently re trapping them in a linear Paul trap, where sympathetic cooling by laser-cooled Be<sup>+</sup> ions reduces the translational temperature of the HCI to the millikelvin regime [10].

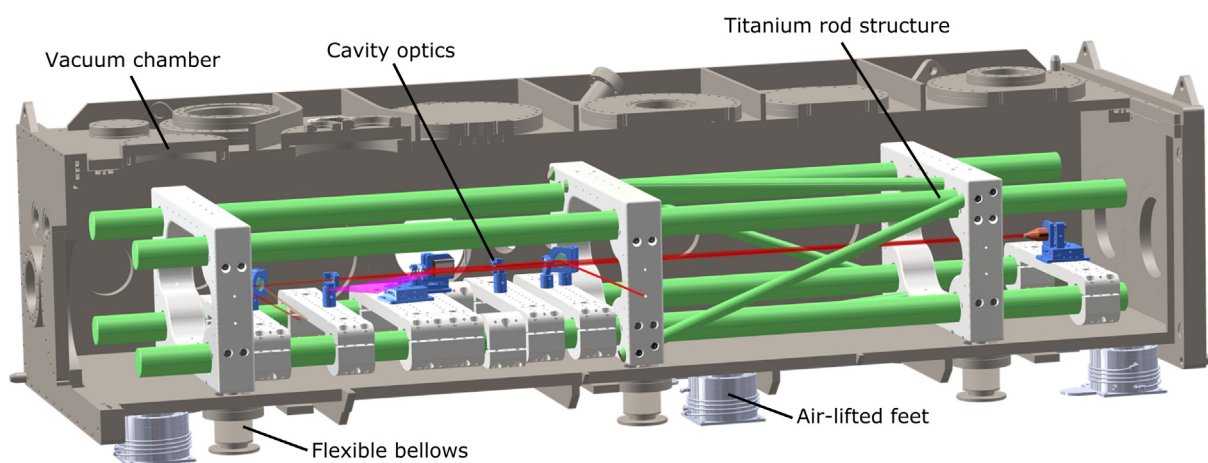
While CryPTEx aims at performing laser spectroscopy in the optical range, the vast majority of electronic transitions in HCI are located in the extreme ultraviolet regime (XUV). These can be accessed using a free-electron laser (FEL) [11], or a synchrotron radiation facility [12], but the coherence of these sources is limited compared to what is achievable with optical lasers. In this project, we aim at building a light source using high-order harmonic generation (HHG) to create a frequency comb in the XUV regime with wavelengths below 100 nm. This method has already been proven to transfer the coherence from infrared (IR) light [13,14], where techniques of radio-frequency/optical phase stabilization are well established, towards the XUV range. We plan to use this XUV frequency comb to perform spectroscopy on trapped and cold HCI at unprecedented accuracy.

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**Fig. 1.** Overview of the femtosecond enhancement cavity. Infrared (IR) pulses are coupled in through the in-coupling mirror (IC) and circulate in the cavity composed of four other high-reflective (HR) mirrors. In one of this mirrors, a shallow grating structure is etched. The inset shows high-order harmonic generation (HHG) inside the tight focus of the cavity, created by the two curved mirrors in the middle (CHR). The high-order harmonics (labeled XUV) propagate collinearly with the IR beam, and are coupled out of the cavity using the minus-first order diffraction of the grating.



**Fig. 2.** Design of the vacuum chamber (dark grey) and rod structure (green and light grey) supporting the mirrors (blue) of the enhancement cavity. The rod structure can be taken out of the vacuum chamber for convenient modification via either the top lid or both smaller side lids. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

## 2. A frequency comb in the ultraviolet

The highly non-linear process of high-order harmonic generation takes place when an atom is exposed to a very intense light field, exhibiting electric field strengths comparable to the intra-atomic Coulomb potential. The process can be described by a three-step model [15]. After tunnel ionization and subsequent propagation of the free electron in the laser field, recollision with the parent ion can result in the emission of odd high-order harmonics of the fundamental radiation. The maximum photon energy which can be reached depends on the ionization potential of the target gas, the peak electric field and the laser wavelength.

One of the important implementations of HHG is time-resolved spectroscopy, where attosecond pulses can be generated [15]. In this field, the intensity which is required for HHG, roughly  $10^{13}$  W/cm<sup>2</sup>, is usually reached by simply focusing a mJ-energy laser pulse to few tens of micrometers spot size. These systems operate at repetitions rates ranging from a few Hz to several kHz.

In this project we are interested in frequency-resolved spectroscopy with a relative precision better than  $10^{-10}$ . To make this possible, we use a frequency comb: a mode-locked femtosecond laser with stabilized carrier envelope offset frequency ( $f_{\text{CEO}}$ ) and repetition rate frequency ( $f_{\text{rep}}$ ). The optical frequency of any comb line is defined by these two radio frequencies and the mode number of the comb line. Since  $f_{\text{CEO}}$  and  $f_{\text{rep}}$  can be readily measured

and locked to a reference source, one only needs to determine the comb tooth number in order to obtain an absolute frequency measurement [16]. The latter is only possible if the spacing between adjacent comb teeth is larger than the linewidth of the transition to be measured. For HCI this means  $f_{\text{rep}}$  needs to be well above 1 MHz, which, when assuming constant average power, comes at the cost of peak intensity.

The lack of power can be compensated using an enhancement cavity, which recycles the IR pulses. With comb line spacings of many MHz, the size of this optical resonator remains suitable for tabletop experiments. The cavity is designed to have a very tight focus to increase the peak intensity and enable HHG. At the focus, a target gas is introduced and an XUV spectrum is generated, containing a copy of the IR frequency comb at each odd multiple of the original frequency [16,13]. Tunability of the individual comb lines will be possible by slightly changing the stabilized repetition frequency of the comb. Since the spectrum will be covering a broad range in the XUV, the light can be used to access many different transitions in a variety of ion species.

## 3. Experimental overview

We use a commercially available Yb-doped fiber laser (FC1000-250 from Menlo Systems) to generate 100 nJ pulses with a duration

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