Frequency metrology using highly charged ions

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Abstract. Due to the scaling laws of relativistic fine structure splitting, many forbidden optical transitions appear within the ground state configurations of highly charged ions (HCI). In some hydrogen-like ions, even the hyperfine splitting of the $1s$ ground state gives rise to optical transitions. Given the very low polarizability of HCI, such laser-accessible transitions are extremely impervious to external perturbations and systematics that limit optical clock performance and arise from AC and DC Stark effects, such as black-body radiation and light shifts. Moreover, AC and DC Zeeman splitting are symmetric due to the much larger relativistic spin-orbit coupling and corresponding fine-structure splitting. Appropriate choice of states or magnetic sub-states with suitable total angular momentum and magnetic quantum numbers can lead to a cancellation of residual quadrupolar shifts. All these properties are very advantageous for the proposed use of HCI forbidden lines as optical frequency standards. Extremely magnified relativistic, quantum electrodynamic, and nuclear size contributions to the binding energies of the optically active electrons make HCI ideal tools for fundamental research, as in proposed studies of a possible time variation of the fine structure constant. Beyond this, HCI that cannot be photoionized by vacuum-ultraviolet photons could also provide frequency standards for future lasers operating in that range.

1. Introduction
Most of the baryonic matter contained in the universe is virialized within large gravitational potential wells of galaxy clusters that shape the intergalactic medium, and therefore appears in ionization states given by the high temperature of that medium ($10^5$ K and above) and the atomic number of the corresponding element [1,2]. Within galaxies, the fraction of ionized hot gas is also the largest one; stars basically contain only highly ionized matter. Reionization of the universe is mostly driven by the energy released by stars, quasi-stellar objects, and active galactic nuclei. Therefore, most elements appear naturally as highly charged ions (HCI) with an unbalanced positive charge. In spite of their importance, for both experimental and theoretical reasons our knowledge of HCI is still rather limited.

Crucially, the electronic structures of HCI, exception made of a few isoelectronic sequences, are not accurately known. Only hydrogenlike and lithiumlike ions have been subject to many studies in the x-ray and vacuum ultraviolet (VUV) domains for their theoretical “simplicity”; also the closed-shell isoelectronic sequences, e. g., heliumlike and neonlike, have been investigated due to their strong abundance in hot plasmas. Besides of the difficulties in production and handling of HCI, which have certainly constrained their study, the main interest of the HCI community has rested for the last three decades, on one hand, in the investigation of quantum electrodynamic properties, relativistic electronic structure and on the other hand in the astrophysically abundant ion species. An introductory review on HCI is given in Ref. [3].
As the key spectroscopic tools available for their study were wavelength-dispersive crystal or grating spectrometers and energy-resolving solid-state detectors, the achievable resolution was very limited in comparison with the much more advanced optical and laser spectroscopy of neutrals and singly charged ions. Due to this, the current status of HCI spectroscopy, from the x ray to the optical range, includes only a few transitions for which the wavelengths are known at the ppm accuracy level, and very few with a just sub-ppm-accuracy. An overview of this field is given in Ref. [4].

1.1. Optical transitions in HCI: Experimental status
Arguably, although the forbidden optical transitions of HCI were, unbeknownst to the astronomers of the 19th century observing solar spectra (cf. Ref. [5]), the first forbidden lines seen in spectroscopy, there is only a small set of HCI optical lines in the spectral atlases. Astrophysical observations have yielded a number of HCI coronal lines for a few abundant elements, such as Fe and Ar. Laboratory work has focused on those and a few other examples, such as the titaniumlike [4-6] and boronlike isoelectronic sequences [4, 8-13]. As for the hyperfine transitions in the ground state of hydrogenlike ions [14,15], they have been observed for $^{209}$Bi$^{52+}$[16], $^{211}$Pb$^{81+}$ [17] ions using an ion storage ring, and for $^{165}$Ho$^{60+}$ [18], $^{185,187}$Re$^{74+}$[19], and $^{203,205}$Tl$^{80+}$ [20] ions with an electron beam ion trap (EBIT) [21,22]. In both setups, the achieved resolution was in the order of 100 ppm. An overview on the different experiments performed with trapped HCI is given in [4]. HCI are mostly produced and studied in the laboratory with EBITs. In those devices, a monoenergetic electron beam produces, traps and excites HCI in any desired charge state of interest up to bare U$^{92+}$ under well-defined conditions [21,22]. At Max-Planck-Institut für Kernphysik (MPIK) we operate a facility with several EBITs dedicated to fundamental physics research and some astrophysical and plasma applications.

As of today, there is still a very large range of totally unexplored HCI. Many isoelectronic sequences with semi-filled electronic subshells never have attracted any strong interest for the reasons mentioned above. In particular, the plethora of forbidden transitions that appear in their ground states has only recently began to be investigated in the detail needed as a prerequisite for laser excitation studies [23,24]. In view of this data scarcity, in the first place systematic studies of the emission spectra of the isoelectronic sequences of interest under electron-impact excitation conditions are needed. For such studies EBITs are very useful: they provide selectable charge states and easily allow the observation of M1 (magnetic dipole allowed) transitions with decay rates in the range of only 30-500 Hz by using broad-band grating spectrometers and cooled charged-coupled cameras with exposure times of less than one hour. However, while finding new lines in the spectrum of a certain charge state is comparatively easy, assigning them to certain transitions is, in most cases, a far more difficult task. Unfortunately, theoretical calculations are very complex and do not yield the accuracy level which would be desirable for reliable line identification. For these reasons, it becomes necessary to obtain additional information by recording spectra along an isoelectronic sequence. Thereby, smooth changes of the line positions and intensities can be observed and provide clues for the identification process. By carrying out such systematic studies, reliable identifications can be gained. The experimental data obtained with an EBIT on, e. g., M1 and very weak E1 lines at level crossings allows for the determination of the level structure and enables the search for transitions with extremely low decay rates. An example is our recent analysis of the optical spectrum of the Ir$^{17+}$ ion [23], a prime candidate for the search of a time variation of the fine structure constant.

1.2. Production of cold HCI and future applications
Albeit HCI production and study experienced a tremendous bonanza with the introduction of EBITs, a key limitation of those devices (and in general of all sources of HCI) is the high translational temperature of the ions trapped in the deep confining potential generated by the negative space charge of the electron beam itself. Values in the range of MK, or hundreds of eV, with heating resulting from the scattering and ionization interactions with electrons, are typical. Since this was an improvement in comparison with plasma sources, those high values did not much hinder x-ray research hitherto. However, up to now they constitute an unsurmountable problem for high-resolution laser spectroscopy.
Recently, the sympathetic cooling of highly charged ions (HCI) [25,26] in a linear RF trap has been demonstrated in our group by L. Schmöger in collaboration with Physikalisch-Technische Bundesanstalt (PTB), thus, opening this broad class of spectroscopic targets to high precision frequency metrology. For this purpose, a cryogenic RF trap, CryPTEx [27-30] had been developed at MPIK in collaboration with Aarhus University. Upgraded devices are now under construction at MPIK in collaboration with the group of P. O. Schmidt for their future use at PTB in the frequency metrology division. With the rapid development of frequency combs in the VUV region and possibly beyond, further applications of highly charged ions as frequency standards at higher photon energies – the typical realm of HCI spectroscopy – are now under consideration, and experiments are being prepared in our group for exploring those.

2. Laser spectroscopy with HCI

While the first photon excitation experiments on HCI were carried out with E1-dipole allowed transitions at 48 eV energies [31], and subsequently at even higher values, first attempts to excite forbidden transitions with lasers within an EBIT showed that a direct detection of the fluorescence from long-lived, laser-excited metastable states decaying through M1-dipole transitions with lifetimes in the order of several milliseconds was possible. Therefore, for two of the forbidden lines that had already been studied with EBITs under electron beam excitation conditions, laser spectroscopic investigations within the EBIT were also performed by utilizing the so-called magnetic trapping mode [32]. This method consists in turning off the trapping electron beam of the EBIT and using its strong magnetic field and the trapping potentials applied to the drift tubes of such a device to keep the ions confined. This mode of operation is equivalent to that of a Penning trap, and has been used to measure the lifetimes of metastable levels with lifetimes ranging from the microsecond to the millisecond range [4,11,12]. The extremely low residual pressure in the cryogenic vacuum of an EBIT (10^{-13} mbar typically) reduces HCI losses due to charge exchange to negligible levels. Furthermore, ion-ion collisions occur at very far distances in the atomic scale, with the result that quenching of metastable levels through collisions is suppressed. The combination of strong confinement and negligible losses allows for the observation of slow radiative decay channels for seconds and minutes, and delivers very accurate lifetime determinations.

2.1. Laser excitation of forbidden transitions

Examples of HCI that have been studied with laser excitation are the boronlike Ar^{13+} ion [33] and the aluminumlike Fe^{13+} ion (Fe XIV) [34], this latter being the source of the long-unexplained “green coronal line” of solar eclipses [5]. For those experiments, a repetitive measurement cycle lasting typically 1 s was used. First, the ions of interest were produced with the electron beam; then, this was turned off and it was waited during a few lifetimes for the decay of the large metastable population populated by the electron beam excitation. A pulsed (8 ns, 100 Hz, 30 mJ, 0.04 cm^{-1} bandwidth) tuneable dye laser then illuminated the ions while a fast shutter closed for ~1 ms the aperture leading to a photomultiplier equipped with a light guide and a filter for the wavelength range of fluorescence. This proved crucial to reduce stray light to acceptable levels. At resonance, and after application of evaporative cooling to the trapped HCI, the laser interaction with a typical ensemble of 10 million trapped ions at a temperature of 20 eV (200 000 K) would generate fluorescence rates of tens per second, well above the dark and stray light count rate. Doppler broadening under these conditions is extremely large, at the level of several GHz. Even with a careful calibration, the resulting experimental wavelength uncertainties are still on the order of hundreds of MHz. Obviously, while needed as a preliminary step, this method is not useful for clock applications. Similar limitations applied elsewhere, and therefore, since more than two decades methods for cooling trapped HCI from the MK level to the mK level have been called for and tried in various settings [35-38].
3. Sympathetic cooling of HCI: A new tool for laser spectroscopy

For direct laser cooling, it is necessary to have fast cycling optical transitions requiring levels of opposite parity and strong overlap near the ground state. In HCI, the next suitable levels are energetically very far from the ground state configuration. Accessing them with optical lasers is, thus, impossible, and cooling has to rely, e.g., on the indirect method of sympathetic cooling. Coulomb interactions between HCI and singly charged ions, which can be directly laser cooled, such as trapped Be⁺ remove thermal energy from co-trapped HCI. The choice of Be⁺ is motivated by the favourable charge-to-mass ratio of 1/9, sufficiently close to that of typical HCI (~1/20 to ~1/3) to allow for simultaneous trapping in a RF linear trap.

3.1. A cryogenic linear RF trap for sympathetic cooling: CryPTEx

CryPTEx (Cryogenic Paul Trap Experiment) was designed to trap HCI [27,28], and for this purpose the foremost issue is guaranteeing an excellent vacuum. Superior ultra-high vacuum performance of existing cryogenic setups in our group has led us to the choice of such a system. The cryogenic environment shown in Fig. 1 has many advantages, not only for the storage and handling of HCI, but also for the trapping and rovibrational cooling of molecular ions; these advantages were very beneficial for the commissioning experiments carried out with CryPTEx at Aarhus University in collaboration with the Drewsen group [29,30]. The device offers 16 ports with optical access to the central RF trap region with the electrodes operating close to 4 K. Cooling laser beams, a photoionization laser, several excitation lasers, the imaging setup, and injection of well-collimated atomic beams and also of buffer gases are needed to perform these types of experiments.

Beryllium atoms are injected in the form of a strongly collimated atomic beam with a cross section of roughly 1 mm². The atomic beam is generated in an oven located in a separated chamber with one intermediate stage of differential pumping. This avoids deposition of beryllium on RF electrode surfaces, and minimizes contamination of the trap chamber. Under normal operation we load an amount of only ~10 mg of pure metal in the oven every year. Most of the evaporated material is collected in a specifically designed, removable housing surrounding the oven, which also includes the first collimating aperture.

A photoionization laser operating at 235 nm crosses the atomic beam at the trap centre and generates the required Be⁺ ions. The ion cooling utilizes a 313 nm laser system based on two telecom fibre lasers with subsequent sum-frequency generation and frequency doubling that delivers up to 700 mW at the cooling wavelength; for most experiments only 2 mW are needed. The laser frequency is stab-
lized with a wavemeter and its intensity with a control system based on a slow regulation with polarizers and a fast regulation with acusto-optic modulators. The laboratory has been temperature stabilized with a home-made control unit which controls the air condition system, reducing the temperature fluctuation to a level below 0.1 K.

For refrigeration, CryPTEx employs a 1 W cryocooler, which brings the temperature of the trap elements and the inner cryogenic shield down to ~4 K, and of the outer cryogenic shield to ~30 K. Turbo-molecular pumps are used in addition to provide a base vacuum in the 10^-9 mbar range. The laser and ion beams enter the differentially refrigerated parts of the system through narrow tunnels for reduction of both thermal radiation from the room temperature vacuum chamber and ballistic residual-gas inflow. Under cryogenic operation, the pressure at the trap centre was estimated to be ~10^-15 mbar by measuring the lifetime of trapped ions (in excess of 1 day) and the heating and decay rates (at ~mHz level) of rotational excitations in molecular ions. In this way, an “effective” black-body radiation temperature of ~7.6 K was determined. For MgH^+ ions, the rovibrational ground state could be the most populated one. This could also be achieved with a novel method in which rovibrational cooling was accelerated by very rare collisions (a few per second) with a tenuous He buffer gas of only ~10^10/cm^3, that is, four to five orders of magnitude lower density than in typical buffer-gas cooling settings, which did not melt the ion crystal. Rovibrational temperature control through conversion of micromotion in heating rate via this rare collision was also achieved [29,30].

The RF trap operates at 4 MHz and has 24 electrodes of 8 mm diameter, of which 12 are used to confine the crystals. Typical trap frequencies are 400 kHz (radial) and 150 kHz (axial) for the Be^+ ion. The ion crystal is imaged with a lens or a microscope objective onto an intensified CCD. Two photomultipliers with appropriate dichroic mirrors and interference filters are used in parallel to register the Be^+ fluorescence rate and the Ar^{13+} fluorescence. A movable fibre mounted on a manipulator is used as a fiducial mark for the alignment of the lasers and the imaging system.

3.2. External ion source: Electron beam ion trap

For the purpose of HCI production, HYPER-EBIT was used as a source of ions for the present experiment. This device, originally designed for high-intensity operation, became available for the present experiment. In the particular example of Ar^{13+}, an electron beam of ~35 mA at an electron-beam energy of 700 eV is used. Under these conditions, the trapped ions typically reach within the trap a translational temperature of 1 MK. The HCI are dumped from the EBIT with a 1 Hz repetition rate with a total kinetic energy of ~6 keV. The charge states of the ions which are transported to the trap through a ~2.5 m long beamline are selected by controlling their time of flight. The HCI are decelerated prior of entering the CryPTEx chamber by application of high voltages pulses during their passage through specially designed electrodes next to it. After overcoming the bias potential of the RF trap setup, the mass-selected HCI bunches enter the RF trap. An electrode is then biased to close the trap axially. The HCI execute an oscillatory motion along the trap axis in its full length of ~100 mm. During this, they interact several times with a continuously cooled Be^+ ion crystal, thereby loosing kinetic energy until they are finally trapped in crystal sites along the RF node, i.e., the trap axis.

3.3. Diagnostics of HCI cooling process

Images captured in exposures of ~1 s duration by the intensified CCD located below the main chamber show the stopping of HCI and their implantation into the pre-existing Coulomb crystal by the appearance of dark, spherical hollows in the crystal axis that are caused by the Coulomb repulsion of the HCI and the Be^+ ions [25,26], as shown in Fig. 2. Ion-ion separations of ~25 µm are typical for Be^+, and about twice as large around an Ar^{13+} ion. We have experimented with different crystal and ion chain configurations, including the one most suitable for application of quantum logic spectroscopy [39] in optical clocks [40], namely one cooling Be^+ ion and one single HCI. For the different setups we have determined the key trap frequencies and carried out different micromotion studies.

The HCI tend to occupy very stable sites in the crystal and show little sensitivity to parameters of the laser cooling which, on the contrary, clearly affect the position and ordering of the surrounding
Be\(^+\) ions. Due to the fact that the RF pseudopotential acts more strongly on the HCI, and to the shielding of electrostatic offset fields, one can presume that their micromotion is even smaller than that of the cooling ions around them. However, this assumption was not used in our preliminary measurements of a conservative upper bound for the HCI temperature of \(~200\) mK. In summary, the present setup in use is capable of transferring HCI at 1 MK from an electromagnetic trap, HYPER-EBIT to another one, CryPTEx, and refrigerate them down to the level of 200 mK and, presumably, even below that value [25,26].

4. Laser spectroscopy of cooled HCI and optical clocks
As a next step, the trapped Ar\(^{13+}\) HCI will be excited with a narrow-band CW laser at the resonance wavelength of the M1 ground-configuration transition \(1s^2 2s^2 2p^2 P_{1/2} \rightarrow 2p^2 P_{3/2}\). This transition has been studied in our group before and is the best characterized M1 transition in any HCI [8-10]. However, the wavelengths obtained in earlier studies still have large uncertainties, which will make a long search necessary. The key issue at this moment is the long lifetime of the excited state (9.7 ms [12]), the small number of trapped ions and the total fluorescence detection efficiency of the system (0.4%). Scanning through the spectral range of interest will require long measurement periods. However, once the exact wavelength is found and calibrated by comparison with a GPS-stabilized commercial frequency comb and a Rb vapour cell, we expect that experiments with the Ar\(^{13+}\) ion and its isotopic varieties will proceed much faster.

4.1. Studies of the time variation of the fine-structure constant \(\alpha\)
Similar experiments with Ir\(^{17+}\) will then follow in order to precisely measure all M1 transitions and other forbidden lines for the determination of the sought-after fine-structure constant-sensitive transitions. Furthermore, other HCI species which have been proposed [41-48] will be investigated. Plans for studies of upper bounds for the Lorentz-invariance in an analogous manner to those of Pruttivarasin et al. [49] but with appropriately chosen HCI, as proposed by Safronova, are also aimed at.

4.2. Clock transitions and other future possibilities
Future work aims at implementation of quantum-logic detection of the clock transition for frequency metrology [39]. This method should provide a far more efficient way of interrogation for the very slow transitions of interest that have been proposed in several recent theoretical works [41-48]. If the present combination of methods is successful, we expect that HCI will provide ultra-stable frequency standards and probes for interesting open questions in physics. Beyond that, the study of the intrinsic properties of their electronic structure, which by virtue of scaling laws can be compared to a virtual microscope for relativistic, quantum electrodynamic, and nuclear size effects will certainly also open new possibilities for atomic physics research.

4.3. Next upgrade: CryPTEx-II
Currently, CryPTEx-II, an upgraded version of the current cryogenic Paul trap with improved stray field and vibration suppression is being setup in collaboration between MPIK and PTB. This platform will be used for the implementation of the quantum logic scheme. The setup will include a miniaturized EBIT, which operates at room temperature and utilizes a magnetic circuit with permanent magnets generating a field of 0.85 T. This much-simplified version should suffice to produce the ions, which are currently the aim of this research. A prototype in operation at MPIK has already generated
beams of, e.g., Xe\textsuperscript{40+} ions. The PTB device is currently being assembled at MPIK. A deceleration beamline similar to that of CryPTEx is also in the design stage at MPIK and will become part of CryPTEx-II at PTB. Commissioning of the complete setup is expected to take place before the end of the year 2016. A twin setup will also be built at MPIK to support further HCI experiments on its site.

5. Perspectives: Vacuum-ultraviolet range and x-ray domain frequency standards

One of the most important advantages of HCI is their stability at high temperatures and also in the presence of high energy photons. Under those conditions, atoms are obviously not stable, and even their inner-shell transitions become broadened by the coupling to the continuum given by the Auger decay. Not so with HCI, which, depending on their charge state, can in principle survive in photon beams of up to 140 keV, in the extreme case of the U\textsuperscript{91+} ion. While E1 x-ray transitions become very broad due to their short (fs range) lifetimes, there are also various electronic configurations that have metastable states of remarkably long lifetimes.

A good example for a possible VUV clock transition is found in the heliumlike O\textsuperscript{6+} ion, with its lowest excited state 1s2s\textsuperscript{3}S\textsubscript{1} decaying to the ground state via a M1 transition at 560 eV (2.2 nm) with a lifetime of \textasciitilde 1 ms [50]. With a transition frequency of \textasciitilde 140 PHz and this lifetime, this atomic resonator possesses a quality factor Q value of approximately 10\textsuperscript{14}. For the M3 decay of the lowest excited state of nickellike \textsuperscript{129,132}Xe\textsuperscript{26+} ions, a transition energy of 1450 eV and a lifetime of \textasciitilde 15 ms were measured with an x-ray microcalorimeter [51]; this yields for Q \approx 5\times10\textsuperscript{15}. With even higher-order forbidden transitions, the corresponding Q values can be even higher, and also higher photon energies can be reached.

In principle, sympathetically cooled HCI will be useful as frequency standards at high photon energies for which until now only crystallographic references are available. Those have a several orders of magnitude lower intrinsic accuracy and stability than electronic transitions in HCI. In principle, HCI could support x-ray frequency measurements with Hz accuracies, if appropriate x-ray sources based on high-harmonic generation would eventually be introduced. Present work at MPIK includes the development of a high-harmonic frequency comb for the VUV range, which should allow exploring the range of electronic transitions beyond the optical, with a hopefully promising scientific harvest.

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