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Laser spectroscopy on trapped highly-charged ions using soft X-rays from FLASH

S W Epp1, J R Crespo López-Urrutia1, G Brenner1, V Mäckel1, P H Mokler1, R Treusch2, M Kuhlmann2, M V Yurkov2, J Feldhaus2, J R Schneider2, M Wellhöfer3, M Martins3, W Wurth3 and J Ullrich1

1Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany
2DESY, D-22607 Hamburg, Germany
3Institut für Experimentalphysik, Universität Hamburg, D-22761 Hamburg, Germany

Abstract. In vast regions of the universe highly charged ions (HCI [1, 2]) are the predominant form of visible matter. Their importance extends to high-temperature terrestrial plasmas, such as those used in fusion research. Yet, accurate prediction of their electronic structure remains a challenge for theory due to the strong electromagnetic field in which the remaining bound electrons dwell. Experimental accuracy has now reached the performance limits of conventional photon spectroscopy in the soft and hard x-ray regions. In this work [3], we report on the resonant laser excitation of the \( ^2S_{1/2} \rightarrow ^2P_{1/2} \) transition of the Li-like Fe\(^{23+} \) ion at 48.6 eV, an energy range hitherto unattainable with powerful lasers. The HCI stored in an electron beam ion trap (EBIT [4]) were resonantly excited by ultra-brilliant radiation generated at the Free electron LASer in Hamburg (FLASH [5]). While yielding a relative statistical error of only \( 2.2 \times 10^{-5} \), and extending laser spectroscopy on HCI from the near ultraviolet [6] to the soft X-ray region, this novel experiment demonstrates immediate potential to push the current limits of precision by orders of magnitude. Such experiments allow to verify predictions of quantum electrodynamics (QED) in a strong field environment where perturbation theory [7, 8] fails. Future EBIT experiments at upcoming x-ray free electron lasers (X-FEL) like the Stanford Linear Coherent Light Source (LCLS) or the European X-FEL will pave the way for laser spectroscopy into the hard x-ray region.

epp@mpi-hd.mpg.de

1. Introduction

HCI are everywhere in the universe: stars and their atmospheres, supernovae, near-stellar clouds, shocks and jets from active galactic nuclei, etc. Their spectral features provide us with direct and unique information on the composition and state of matter of even the most distant known objects. Therefore, huge efforts have been undertaken to measure emission lines of astrophysical objects with increasing resolution, culminating in a series of new satellite missions in this decade. HCI up to even hydrogen-like uranium have become available in a few earthbound laboratories, either by accelerator techniques [9], in ion storage rings [10] or in EBITs [11,12,13]. Increasing research efforts in theory and experiment to investigate their structure have yielded tremendous progress on both sides [14]. Beyond their diagnostic importance in plasma physics and astrophysics, HCI provide an ideal testing ground for forefront atomic structure theory, with extensive fundamental implications. So does the leading one-photon exchange contributions from bound state QED not only scale with about the fourth...
power of the nuclear charge $Z$ (thereby increasing their relative contribution to the respective binding energies with $Z^2$) but, moreover, cannot be calculated by perturbation theory, i.e., using a rapidly converging series expansion, since the expansion parameter $Z \alpha$, with $\alpha = 1/137$ being the fine-structure constant, approaches almost unity for uranium. Thus, QED, the most precise theory in physics and the foundation of any quantum field theory, including the Standard Model, can be explored in a unique regime not accessible by any other means, namely in intense electromagnetic fields reaching electric and magnetic field strengths of up to $10^{12}$ V/m and $10^4$ T, as at the surface of a uranium nucleus.

Secondly, HCI allow control of the electronic complexity by selecting the number of bound electrons. Exploring hydrogen-, helium-, or lithium-like systems with the same nuclear charge is one way of varying electron-electron correlation contributions in the system. Relativistic contributions scale to the binding energy with $Z^4$ and, hence, are strongly boosted. Transition probabilities show an even stronger $Z$ dependence, like the M1 transition matrix elements growing with $Z^{11}$. Nuclear size contributions scaling with approximately $Z^2$ become accessible using laser or conventional optical spectroscopy of the hyperfine structure [6,12]. Relativistic nuclear recoil effects have been investigated in unprecedented detail by accurate measurements of isotopic shifts in forbidden transitions of medium-Z HCI [15]. Furthermore, several schemes to study parity non-conservation (PNC) effects or drifts [16] in $\alpha$ by systematically tuning $Z$ and nuclear mass $A$ along certain isoelectronic sequences have been proposed.

Spectroscopy of neutral atoms and molecules was revolutionized by the advent of lasers, with impressive examples of accuracy, like the $\Delta\nu/\nu = 1.8 \cdot 10^{-14}$ recently demonstrated in measurements of the absolute $1S-2S$ transition frequency in atomic hydrogen [17], an improvement compared to conventional spectroscopy by several orders of magnitude. So far, laser techniques could only be applied to HCI in the visible and near ultraviolet spectral range in exceptional cases, such as hyperfine splitting of heavy hydrogen-like ions [6] and fine structure splitting of light ions [18] in which the electronic structure of the HCI allows for an optical transition. Photon emission by HCI is naturally most intense in the x-ray region. The absence of laser spectroscopic data in this region was a consequence of the complete lack of intense x-ray laser sources and of the fact that HCI were only available as very tenuous beams, with densities much below the $10^{10}$ ions/cm$^3$ achieved here with an EBIT. This lack of appropriate lasers meant that x-ray wavelength measurements for HCI had to use conventional wavelength or energy dispersive methods, as e.g. crystal or grating x-ray spectrometers, or solid state detectors. Despite intense efforts invested in instrumentation development over several decades, their absolute limitations with regard to accuracy, stability, and efficiency remained an insurmountable barrier to further experimental progress. As a consequence, the most accurate wavelength measurements in HCI with $Z > 20$ did not exceed 15 ppm [19], systematically limited by instrumental resolving power and statistics. In this aspect, the situation resembled the status of optical spectroscopy prior to the availability of tunable lasers.

Here we report on the first experimental results obtained through the successful combination of two advanced technologies: FLASH [5] (Free electron LASer in Hamburg), the most intense soft X-ray laser in the world, operated at DESY, and a state-of-the-art EBIT, developed at the Max-Planck-Institute for Nuclear Physics in Heidelberg, to efficiently produce and store HCI, which serve as a target. The goal of these experiments [3] is to perform resonant fluorescence laser spectroscopy on ions belonging to the Li I isoelectronic sequence, starting with the transition between the $1s^22p^2{}^3P_{1/2}$ and $1s^22s^2{}^3S_{1/2}$ states (Fig. 1b) for nuclear charges between $Z = 20$ and $Z = 92$. The transition energies lie between about 40 eV and 280 eV, well within the specifications of FLASH, which presently is capable of delivering photons of energies up to 90 eV (with 200 eV becoming available by 2007). Li-like ions, first observed in solar flares [20], are particularly interesting among HCI for strong field QED studies. They have attracted growing experimental and theoretical attention [7-9,13-14,20-28]. The relative QED contributions to the total transition energy summing to 1% ($Z = 20$) or even 15% ($Z = 92$) are exceedingly large.
**Figure 1.** (Colour online) (a) Ions (green) are produced by successive electron impact ionization, induced by an energetic electron beam passing through the trap. By applying appropriate potentials to the three cylindrical trap electrodes (orange), the ions are confined longitudinally to a volume of 50 mm length. The negative space charge potential of the 450 mA electron beam (red), compressed to 50 µm in the trap region by a 6 T coaxial magnetic field B (brown), is able to confine the ion cloud in the radial direction to nearly that diameter. Soft x-rays (light blue) from FLASH enter the trap as pulse-trains with a 5 Hz frequency. Each train consists out of 30 single pulses spaced by 1 µs (upper inset). The photon pulses (lower inset) excite the Li-like Fe\(^{23+}\) ions according to Figure 1b. Resonant fluorescence radiation also emitted in radial direction can be recorded. (b) Excitation scheme from the ground level of a Li-like system by resonant laser radiation. (c) Typical example of the spectral density distribution of FLASH pulses before passing the monochromator.

1. **Experiment**

In our proof-of-principle experiment [3] we measured the above mentioned transition in the Li-like iron ion, Fe\(^{23+}\), according to the scheme illustrated in Figure 1b. Due to the E1 character of this transition, the lifetime of the excited level is only 0.55 ns. Therefore, its excitation by a resonant laser will result in immediate emission of fluorescence radiation, in particular also in the direction perpendicular to the laser beam, towards the collecting mirrors. The idea is now to measure the intensity of this fluorescence as a function of the wavelength by scanning the laser over the line at roughly 48.6 eV, or 25.5 nm.

As depicted in Figure 1a, Li-like Fe\(^{23+}\) ions (Z = 26) were produced in the Heidelberg EBIT by successive electron impact ionization of Fe by means of an energetic electron beam. Fe atoms are brought into the trap in form of a tenuous Fe(CO)\(_5\) molecular beam. These molecules are dissociated by electron impact in the trap. The ions in an EBIT [4, 11] are confined longitudinally by appropriate potentials applied at the cylindrical trap electrodes (Fig. 1a). In the radial direction, trapping is caused by the negative space charge potential generated by the 450 mA electron beam, which is compressed to a 50 µm diameter by a 6 T coaxial magnetic field. In order to optimize the yield of Fe\(^{23+}\) ions, the electron was accelerated to approximately 5.1 keV by applying a potential between the central trap electrode and the cathode (Fig. 1a). This choice was due to the presence of a strong dielectronic resonance [28,29] at that beam energy. This resonance —essentially an inverse Auger process— recombines the abundantly produced He-like Fe\(^{24+}\) ions very efficiently into the Li-like charge state, thus maximizing it. The beam energy was optimized with two Ge-detectors monitoring the yield of the x-ray Kα photons emitted by the relaxation of the recombined ions (Fig. 2). The detectors also register x-ray emission following direct excitation of the ions by electron impact, and radiative recombination of beam electrons with the HCI (i.e., the inverse process of photoionization). The trapped HCI form under the present conditions, a cloud of 50 mm length with a diameter of roughly 200-300 µm at a density of about 10\(^{10}\) ions/cm\(^3\).
Figure 2. (Colour online) Transverse cross section of the trap region. Fluorescence photons are collected by means of cylindrically shaped mirrors working at a grazing incidence angle of roughly 10°, and focussed onto a microchannel-plate detector (MCP). A grazing incidence flat field grating spectrometer monitors soft x-rays in the range 30 to 100 eV. X-rays above 1 keV are measured by two Ge detectors. One radial port is used for the injection of the Fe atomic beam.

The ion cloud was brought to overlap with the pulsed laser radiation of FLASH, which had a comparable diameter (Fig. 1a, Fig. 1a lower inset). The FLASH beam (light blue in Fig. 1a) consists of pulse-trains of 30 single photon pulses (upper inset Fig. 1a) with a 1 µs separation between them. The trains are produced at a 5 Hz repetition rate, yielding to overall 150 laser pulses/s. Each pulse contains approximately $2 \cdot 10^{12}$ photons. Total flux amounts to roughly $3 \cdot 10^{14}$ photons/s. The pulse length of a single pulse is 20-80 fs. As a consequence of the production mechanism of the FLASH pulses, which is self-amplified spontaneous emission (SASE), the pulses have a bandwidth of roughly $E/\Delta E \approx 50$ to 200, for the average of a pulse train (Fig. 1c). With those parameters the present measurement would not be feasible, since the laser beam is too undefined in its energy. Hence, our experiment uses a scanning plane grating monochromator (PGM) located at the PG2 beamline to reduce the bandwidth of the laser radiation. Of course, this reduces also the total flux of photons by 2 orders of magnitude or more, depending on the desired resolution. With a slit width of 200 µm at the exit slit of the monochromator (Fig 1a), the beam had a cross section of $(200 \times 200) \, \mu m^2$ leading to a resolution $E/\Delta E$ of about 2,000 at 48.6 eV photon energy in our setup. With other settings a resolution of 30,000 could be achieved. In order to facilitate the initial tests and adjustments, the value of 2,000 was chosen as a compromise between photon flux and resolution. Technically, the exit slit was set to 700 µm, but since the ion cloud was only 200-300 µm in diameter it acts effectively like a narrower second slit. After passing the trap region, the photon beam is dumped onto the electron gun cathode (Fig. 1a). A photoemission signal derived from that electrode is used for timing purposes as well as for the determination of the relative pulse intensities.

The fluorescence photons emitted perpendicularly to the photon beam by the decay of the excited state (Fig. 1b) are collected by two cylindrically grazing incidence mirrors and focused onto the surface of a microchannel plate (MCP), see Figure 2. The mirrors are manufactured by bending thin silicon plates with a root-mean-square surface roughness of 0.5 nm. They are attached to one of the seven radial bores of the magnetic coil support structure, providing optical access to the trap region. The total detection probability (detection efficiency times solid angle) is estimated at $10^{-4}$. Resonant fluorescence photons arriving at the MCP are recorded in coincidence with the pulsed FEL photon beam as a function of the photon energy selected by the monochromator. Each FEL pulse starts a time window of 1 µs. Photons arriving at the detector within this window are registered, as well as
their arrival time in relation to the FEL pulse. Since the resonance was expected to be around 48.6 eV, the photon energy was repeatedly scanned between 48.53 eV and 48.71 eV in 5 meV wide steps, with 3 s integration time on each data point for every scan. True coincidences appear in an isolated region in the two-dimensional energy-versus-time representation as shown in Figure 3, with the true coincidences seen in the upper part of the picture in a linear colour scale.

![Figure 3](image.png)

**Figure 3.** (Colour online) $^2S_{1/2}$ to $^2P_{1/2}$ resonance data. 2D plot (smoothed, 5 meV × 0.016 μs binning) of the fluorescence signal as a function of the photon arrival time relative to the FEL pulse (left y-axis, absolute offset of time scale arbitrary) versus photon energy (x-axis). The total measuring time was 1987 seconds. The resonance contains 370 true counts over a 5σ interval after baseline subtraction. Inset: Projection of the 2D representation onto the photon energy axis yielding the number of photons (white squares) per 5 meV photon energy interval (right y-axis) as a function of the photon energy (see text). Red line: Gaussian fit to the data.

2. Results and discussion

Many tests were performed to ensure that the detected photons are emitted from the desired transition in Li-like iron. (i) Displacing the whole EBIT with sub mm precision transversally to the photon beam guaranteed optimal overlap and, at the same time, verified the overall extensions of both the ion cloud as well as of the FEL beam. As expected, the signal disappeared when the overlap was missing. (ii) Switching off the injection of the Fe(CO)$_5$ beam into the trap resulted in a complete loss of true coincidences, demonstrating that no other ions than Fe, C or O possibly trapped in the EBIT could contribute to the measured signal. Additionally, background from photons scattered on the electron beam or on the trap electrodes is ruled out in this way. (iii) Changing the trap potential to expel the positive ions out of its volume made the signal disappear as well, indicating that no neutral atoms or molecules can contribute in any way to it. This all was doubly checked by the identification of the lines recorded by our grating spectrometer (Fig. 2 upper left). This instrument allows one to verify that in the range scanned by the beamline monochromator the line of interest was not blended with any other. Conventional analysis of these spectra, completely independent from FLASH and our laser spectroscopic approach, showed a resolving power $E/\Delta E\approx 500$ for this instrument and yielded a preliminary value for the wavelength of the Li-like transition under study of $(25.5121\pm 0.0004)$ nm.
This is in very good agreement with the most accurate published result [25] of (25.5113±0.0005) nm, or (48.5997±0.0009) eV.

In Figure 3 the two dimensional energy-versus-time spectra as well as the projection of the fluorescence signal onto the photon energy axis are displayed after a measuring time of only 1987 seconds. Clearly visible is a pronounced resonance signal containing about 370 true counts. Out of a Gaussian fit of the data (red curve in Fig. 3) we obtain a transition energy of (48.6127 ± 0.0011 stat. ± 0.0150 syst.) eV. So far, the error bar of this value is completely dominated by the preliminary calibration uncertainty (± 0.0150 eV) of the beamline monochromator. It will be reduced by an adequately accurate calibration of that instrument in the near future.

Therefore, our laser spectroscopic result cannot yet compete in terms of absolute accuracy with those results reported from the most precise measurements performed on Li-like heavy ions [19,26] (20 ppm). Nevertheless, with a statistical error of 23 ppm reached in half an hour measurement time, our result challenges them in terms of statistical accuracy. It also surpasses the statistical accuracy of the most precise beam-foil measurements [22] (48 ppm) by about a factor of two, and that of the most accurate recent EBIT measurement [13] for U⁸⁹⁺ (36 ppm statistical) by a factor of about 1.7 as well. For comparison, the estimated accuracy of theoretical values is around 80 ppm for ions in this Z range.

3. Conclusions and outlook

The potential of the novel method demonstrated here becomes obvious when considering an available monochromator resolving power up to 200,000 achieved at third-generation synchrotrons. The projected gain in accuracy is stupendous. Sub-ppm precision will predictably become possible when the FLASH facility reaches its full capabilities. Moreover, direct resonant fluorescence spectroscopy in the keV regime with the EBIT will be enabled with the commissioning of free electron x-ray lasers like the Stanford Linear Coherent Light Source (LCLS), scheduled for the year 2008, or the European X-FEL in 2013. Those facilities will push the current limits in precision spectroscopy of HCl, with expected improvements by factors of 10 to 1,000 for any transition photon energies up to tens of keV.

With shorter wavelengths becoming available, photoionization studies for HCl will also be feasible. Two methods will be applied in connection with the EBIT: (i) to direct the photon beam to the ions trapped in it, extracting them after a certain interaction time from the trap and analyzing their charge state, to obtain total cross sections, and (ii) extracting HCI beams and crossing them with the FEL radiation in a reaction microscope [30] to determine differential ionization cross sections, which are urgently needed for comparison with theory.

In addition to these advantages, the ultra-fast, 20 fs time structure of the FEL radiation sources (with even attosecond pulse lengths under discussion) will make precise lifetime measurements in a hitherto inaccessible time scale possible. Lifetime measurements reveal aspects of the electronic structure which are not easily investigated by measuring transition energies. While these are primarily determined by the radial properties of an individual wavefunction, lifetimes are sensitive to the entire spatial parts and correlation terms in the overlap of the two wavefunctions in the initial and final state. Steep scaling laws as a function of Z for the lifetimes of levels depopulated by multipole transitions (M1, E2, 2E1, M2...) facilitates those studies with HCI. Recent lifetime measurements in systems with relativistic bound electrons [31] have become sensitive to certain fundamental aspects of the electromagnetic interaction, such as the electron anomalous magnetic moment (EAMM), which until now could not be probed due to the general lack of accuracy of the data.

The present experiment is an important and encouraging step in these directions, and shows for the first time a practical realization of laser spectroscopy in this spectral range, and the wide range of possibilities opened up to research by soft x-ray lasers of high brilliance such as FLASH.

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