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Guest Editor's Notes on the "Atoms" Special Issue on "Perspectives of Atomic Physics with Trapped Highly Charged Ions"

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Abstract: The study of highly charged ions (HCI) was pursued first at Uppsala (Sweden), by Edlén and Tyrén in the 1930s. Their work led to the recognition that the solar corona is populated by such ions, an insight which forced massive paradigm changes in solar physics. Plasmas aiming at controlled fusion in the laboratory, laser-produced plasmas, foil-excited swift ion beams, and electron beam ion traps have all pushed the envelope in the production of HCI. However, while there are competitive aspects in the race for higher ion charge states, the real interest lies in the very many physics topics that can be studied in these ions. Out of this rich field, the Special Issue concentrates on atomic physics studies that investigate highly charged ions produced, maintained, and/or manipulated in ion traps. There have been excellent achievements in the field in the past, and including fairly recent work, they have been described by their authors at conferences and in the appropriate journals. The present article attempts an overview over current lines of development, some of which are expanded upon in this Special Issue.

Keywords: ion trapping; atomic spectra; atomic lifetime measurement; highly charged ions

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1. Introduction

Spectroscopy of highly charged ions has a history of about 90 years by now. The production of highly charged ions became possible when vacuum technology had progressed far enough to prevent premature discharges between high voltage electrodes in vacuum vessels. The combination of high voltage (available from influence electricity generators since about a century and a half by then), high voltage capacitors and coils permitted the experimenters to increase the pulse power of the intended vacuum sparks and to shape the discharge pulse. In the 1920s, Manne Siegbahn intended to push optical spectroscopy towards the X-ray range (in order to connect the two spectral ranges) and tasked the young researcher Bengt Edlén with testing a prototype grazing-incidence spectrograph. Edlén explored the performance envelope of this type of device. By the early 1930s, he had studied the extreme ultraviolet (EUV) spectrum of hydrogenlike lithium ions with an accuracy at the brink of detecting QED effects ("invented" only a few years later on the basis of Dirac's relativistic treatment of the electron). By the late 1930s, Edlén had trained his vacuum spark so that he could observe the spectrum of the Mg-like ion of cobalt (Co XVI), a record high charge state at the time [1]. Edlén and Tyrén discovered satellite lines to the X-ray diagram lines of few-electron ions [2], that is, the presence of outer electrons that play a spectator role to transitions in the EUV and X-ray ranges. The detailed study of highly charged ions enabled Edlén a few years later (encouraged by a suggestion by W. Grotrian) to demonstrate that the wave numbers of the then mysterious prominent coronal lines

coincided with fine structure intervals of highly charged ions of Ca and Fe [3]. Such highly charged ions could not be produced at the 5500 K temperature (0.5 eV) of the Sun's photosphere but required temperatures in the MK range (200 eV and higher), forcing a new perception of the solar environment. The direct excitation and observation of the coronal lines (electric-dipole forbidden transitions in the ground configurations of heavy ions, with wavelengths in the visible range) succeeded almost another half a century later in the low density environment of ion traps, with accurate wavelength measurements eventually reported from an electron beam ion trap (see below) [4].

The sun was fully recognized as a strong source of X-ray and EUV emission only when, in the 1960s, rockets began to lift spectrographs above Earth's atmosphere. At the same time on the ground, plasma physics began to try for achieving controlled nuclear fusion, especially by discharges of the tokamak type, in devices of increasingly larger sizes. The diagnostics of the plasma inside depends largely on spectroscopy and the simple scaling rules of isoelectronic sequences, with the recognition of spectral features from highly charged ion species immediately providing a tool for temperature estimates. Fusion experiments and solar corona research concern low-density plasmas, to which I will return shortly. When the laser was invented and developed towards higher power in the 1970s, one field of interest became the behavior of matter at high energy density. Of course, spectroscopists saw a chance to study the spectra of highly charged ions in such laser-produced plasmas. Na-like ions of rare earth elements, and Ni-like spectra of elements up to uranium have been reported from the laser-produced plasmas (LPP). However, LPP is a high-density plasma that provides so many electrons that the recombination processes prevent the production of even higher charge state ions. Moreover, the plasma is rapidly expanding, which limits the accuracy of wavelength determinations. Although many such measurements have been tried by the LPP technique, those at the highest charge states have since become suspect of underestimated systematic error.

A competing technique, beam-foil spectroscopy (originally developed by Kay and Bashkin [5–7], for more recent descriptions and further references, see [8,9]), employs a beam of fast ions (in the keV to GeV energy range) and passes it through a thin foil target. Ion charge states up to bare uranium have been reached by this technique, but the high ion velocity causes problems for accurate wavelength measurements. The observation in the laboratory rest frame of such rapidly moving ions is helped by the directionality of the ion beam; however, the exact angle between ion trajectory and observation direction is important for correction of the (first order) Doppler shift, and knowledge of the ion velocity is warranted for the time dilation effect (second-order Doppler shift). Moreover, the finite angle of observation causes Doppler broadening (via the range of observation angles), all of which matter at the high level of accuracy wanted in meaningful measurements that nowadays aim at testing QED contributions of higher order. The excitation of the fast ions takes place in a high-density environment (the electron density of the solid-state foil material), while the observation happens in the high vacuum environment of the vacuum vessel after the ions have left the exciter medium; hence, further interactions of the fast ions with the environment or of the emitted photons with absorbing material are prevented. Beam-foil spectroscopy has the enormous advantage of inherent time resolution that permits the suppression of emissions from the vast multitude of core-excited ions (most of which are extremely short-lived) by simply shifting the observation zone to a few micrometers away from the foil, where the fastest decays have already died out. It is possible to follow the ion beam and thus line out the emission signal along the beam trajectory; the resulting decay curve (for a given wavelength) can be interpreted in terms of upper level lifetimes. Thus, beam-foil spectroscopy is unmatched as a technique that gives access to such decay properties as the level decay rates of a vast amount of levels in ions of any element (with isotope selectivity) and charge state, for lifetimes in the range from a few picoseconds (achieved by simple mechanical displacement on the micrometer scale and limited by practical focusing restrictions) to hundreds of nanoseconds (limited by the vacuum vessel size in an ion accelerator environment and by signal statistics). However, many transition rates scale steeply with the ion charge, and the associated lifetimes of levels with only electric-dipole forbidden decays may be too long for the beam-foil technique (with straight ion

beams) for exactly that range of nuclear charge Z (and relatively low charge states) that is of interest in fusion experiments or in astrophysics. In this case, it is necessary to measure atomic level lifetimes in ion traps that may include heavy-ion storage rings, which basically are ion traps that bend the trajectories of fast ion beams into closed loops. Ion traps also play a role in precision spectroscopy, because the storage of ion samples at rather low velocities can minimize Doppler shifts and Doppler broadening. One should, however, keep in mind the very different particle density in the excitation that fast ions experience in foils *versus* the low particle density necessary for working with ion traps, which relates to very different appearances of the associated spectra [10].

Editorial note: Lines of development in studies of trapped highly charged ions are the focus of the present Special Issue. Another special issue, on more general topics of achievements in the spectroscopy of highly charged ions, has concurrently been published by the Journal of Physics [11]. There is, of course, some overlap, and I admit that I have asked some of their authors for contributions to this issue, before I learned of the competing Special Issue project of the other journal. References below and in the contributions to this Special Issue will bridge the gap and provide pointers to the interwoven physics projects.

2. Ion Traps

There are traps that by voltage switching capture ions prepared and excited externally [12,13], and there are experiments in which the ions are produced and/or excited inside the device. There are traps designed to control or study the motion of the trapped ions in fine detail ("precision traps"), and there are strikingly simple trapping geometries that 'work', although the design at first glance may seem far away from and hardly related to the textbook examples (from paperclip electrodes to sets of collinear tubes that form nested Penning traps, and so on). However, this is not the place to discuss the design varieties. In the work with multiply charged ions, the electron beam ion trap (EBIT) has become a workhorse of the community (a historical introduction and many applications are represented in the proceedings of a dedicated conference [14]). In an EBIT, a Penning trap (consisting of three collinear drift tubes on different electric potentials inside a strong magnetic field of the same orientation) is combined with a high-density electron beam that serves several functions. The electron beam produces ions from the residual (or leaked-in) gas atoms; since the ions are being trapped, they can be further hit by the electrons of the beam and be ionized in a stepwise fashion as long as the collision energy exceeds the ionization potential. The trapped ions form an ion cloud (because of the trapping fields), but repel each other. The second purpose of the electron beam is to balance and, actually, over-compensate this space charge of the ion cloud and attract the ions to the electron beam volume. That effect increases the target areal density and thus ensures a quadratic dependence of the optical emission of the ions on the electron beam current.

The EBIT originally was an offspring of an electron beam ion source (EBIS) in which, however, a meter-long electron beam suffered from instabilities; the much more compact EBIT proved much more stable and easy to handle [15,16]. Late in 1986, the EBIT group at Livermore noted the first X-ray signal from highly charged Xe ions in their device [14]. The EBIT concept and development have spread from Livermore to many other laboratories since, while the Livermore EBIT group is looking forward to celebrating the 30th anniversary of the spectroscopy of trapped highly charged ions in 2016. The inception of EBIT happened at a time when X-ray laser development (for example, preparing for resonant photo-pumping between several ion species) ran into problems and urgently needed accurate atomic data in the soft-x-ray range. In the 1990s, extensions to higher voltages were explored. For this, the original EBIT-I was supplemented with a major booster for the electron beam energy. The new configuration was dubbed SuperEBIT and, at electron beam energies beyond 200 keV, produced uranium ions (Z=92) up to the bare nuclei [17–19]. Laboratories in Japan, Germany, and China followed with their own high-voltage EBIT designs. However, high voltage equipment is difficult to handle and to maintain. Arcing and sparking during the training of such a machine can damage the equipment and associated electronics. With limited resources, one soon realizes

that higher voltage carries a higher technical risk, and thus only relatively few experiments have been executed at EBITs running a high voltage in excess of, say, 120 kV, and almost all of them happened at Livermore. The vast majority of EBIT measurements anywhere have been done within the performance envelope of the original EBIT-I. In fact, while an EBIT can produce highly charged ions, many of its scientific targets in plasma and astrophysics concern low charge state ions, and therefore various smaller EBITs have been developed, nowadays often with permanent magnets or with (liquid-nitrogen cooled) high-temperature superconducting coils instead of the classical superconductors and multi-stage cryogenics (and expensive liquid helium) used in the early and high performance EBITs. The small machines typically operate at electron beam energies below 10 keV. However, it is not easy to form and transport a tightly controlled electron beam from an electron gun through a set of drift tubes and then to a collector. Interestingly, the composite design of SuperEBIT in which the electron gun and drift tube potentials are individually adjustable turned out not only beneficial for high energy electron beams, but it also enabled operations at low electron beam energies. The beam then is formed and transported to the trap at a technically advantageous energy in the keV range and is decelerated to a lower energy only near the trap volume. Thus, SuperEBIT has been capable of operating below 100 eV from early on, an achievement duplicated elsewhere only many years later.

As mentioned before, the electron beam energy determines the maximum charge state of the ions in the trap. A low electron beam energy thus enables the selection of a low maximum charge state, and a controlled variation that adds one charge state at a time to a spectrum excited. The contribution by Joel Clementson, Tomas Lennartson and Peter Beiersdorfer to this special issue [25] pushes the low electron beam energy range even further down (this time for the EBIT-I device), to about 30 eV. At such low energies, the achievable electron beam current is low, too, but the excitation cross sections are favorably large so that the photon signal can still be sufficient for statistically meaningful data.

2.1. Complex Spectra

Of course, spectra of low charge state ions have been measured since about a century. One may wonder what an EBIT can do on those spectra that have been obtained from classical light sources and often studied with high spectral resolution and excellent accuracy. A major parameter of interest is the particle (electron) density in the light source. The electron beam in an EBIT features an electron density on the order of 10^{11} cm^{-3}. This is the lowest among laboratory light sources. A tokamak fusion experiment operates at a density on the order of 10^{14} cm^{-3} and is still considered a low-density plasma; planetary nebulae in outer space feature densities on the order of 10^{8} cm^{-3}. The electron density matters for the level populations (which, in turn, result in specific relative line intensities); at very low densities the collision rates are low compared to all radiative rates, and the ions can return to their ground state before the next excitation happens. At high density, collision rates dominate over radiative rates, and higher levels (including multiply excited states) can be reached via multi-step excitation. Beyond this point of principle, obtaining spectra in an EBIT under controlled conditions that help to foresee what may show in a low-density plasma fusion device, there is a matter of practicality. The present-generation fusion reactor designs incorporate tungsten surfaces in areas that will experience a particularly high heat load and risk of sputtering. Hence, plasma diagnostics is interested in observing the plasma near these surfaces where low-charge state ions of tungsten are expected to show. The spectra of these ions are known to be very rich in lines. At the same time such complex spectra are difficult to unravel, because atomic structure calculations are challenged to provide sufficiently accurate indicators. Almost a decade ago, Marius Jonas Vilkas has calculated the level structure of W^{7+} to W^{15+} [20] by using the Multi-Reference Møller–Plesset (MR-MP) approach developed by Yasuyuki Ishikawa and his group.

The many hundred levels per ion (of only the lowest configurations) that Vilkas reported may arguably represent the best computational results at the time. However, for a manageable comparison to experimental data (and for preparing for the plasma diagnostic challenges of fusion plasma devices

such as ITER, see [21,22]) one needs to estimate the relative intensities of the thousands of computed transitions to filter the spectral lines that matter in practice from the many more that may constitute a pseudo-continuum background. Typical programs that handle collisional-radiative models, such as the Flexible Atomic Code (FAC) [23], are less accurate in terms of high-quality level energy computations than MR-MP. One would want to feed the accurate energies from the most accurate computations to the spectral model, but preferably in a somewhat automatic procedure, because there are hundreds of level energies that need to be correctly transplanted. This has not yet been done routinely for the complex tungsten spectra, but recent spectral modeling has been based on increasingly more massive atomic structure calculations.

While reliable spectral synthesis and modeling have not yet reached their accurate goal, EBIT offers a practical tool that is not available (in this rigor) from traditional light sources. In an EBIT, the electron beam energy limits the successive ionization to the highest charge state whose ionization potential can no longer be overcome. By appropriately small beam energy increments, one can track the appearance of new lines and associate them with specific ion charge states, and all that under low-density conditions. This technique has been exercised before especially for iron [24], and it has now been employed by Clementson *et al.* for tungsten, as is described in their contribution to this Special Issue [25]. (Of course, electron beam ion traps are not the only tools used in this research; this journal carries a Special Issue on Atomic Data for Tungsten [26] which describes also very different research methods.)

The same technique has been employed repeatedly with tungsten at the Berlin EBIT, more than a decade ago, including a study at fairly low electron beam energies [27]. At that time, a goal was to produce the Pm-like spectrum of the ion W^{13+}, for which earlier computations had suggested that the 5s–5p transitions might be as prominent as the $\Delta n = 0$ resonance lines in alkali-like ion spectra. Though lines were seen in the Berlin EUV spectra, none of them showed as prominently as had been hoped for, and assignments had to wait for suitable atomic structure calculations. Some of those computations, however, indicated that according to the computed level structure, the 5p levels would not be particularly highly populated either directly or via cascades except possibly in much more highly charged ions along the isoelectronic sequence [28,29]. To my knowledge, no one doubts that the Berlin group may well have observed EUV lines belonging to the Pm-like spectrum of tungsten, except for a recent paper from Shanghai [30]; the author list of this new paper includes one of the Berlin paper authors who so far had insisted that all was well. However, the Vilkas computations [28] have long since indicated that the prominent decays of the 5p levels proposed decades before would not materialize in reality. Even worse, recent radiative-collisional modeling efforts by Kobayashi *et al.* [31] on Pm-like Bi, an element ($Z = 83$) closer to the range of ion charges in which the 5p levels may actually be the lowest of their symmetry and thus be favored by cascade repopulation, have found a population trap, that is a level of relatively long lifetime (low decay rate) in which such cascades would end rather than proceeding to the 5p levels. Therefore, the 5s–5p transitions in Pm-like ions of elements that are not of very high nuclear charge should be considered not just as "not favored", but actually as "disfavored".

Up to this point, the story had seemed to end with indirect evidence. However, very recently, the Heidelberg EBIT group has addressed Pm-like heavy ions again, this time with four elements (Re through Pt, Z=75–78), and combining spectral observations with modeling [32]. Their model calculations predict that the $4f^{14}$ 5s–5p transitions should be very weak at best (and correspondingly weak candidate features are observed in the spectra), whereas a number of $4f^{13}$ $5s^2$–5s5p transitions should appear more strongly (and there are suitable groups of candidate lines). This corroborates conclusions from much earlier beam-foil spectroscopic experiments in which it was found that even with that efficient excitation process 5s–5p lines in Pm-like spectra were not readily produced (the measurements having yielded no more than hints [33]), whereas the observation of $4f^{14}$ $5s^2$–5s5p transitions (in the neighboring Sm isoelectronic sequence) appeared to be more easily achieved [34]. Consequently, one should no longer chase the "ghosts" , the $4f^{14}$ 5s–5p lines, in elements much lighter

than uranium ($Z = 92$), but perhaps try to verify the conjectured population trap by experiment and measure the radiative lifetime of the associated level as a test of atomic structure calculations and radiative-collisional modeling. Electron beam ion traps should be useful tools for that quest.

2.2. Active Interrogation

The fact that EBIT is still an operative ion trap even without the electron beam has many benefits: the excitation can be modulated and even switched off. When the hefty shower of energetic electrons ends, the ions of the trapped ion cloud continue with their radiative decays. By electronic timing, one can record decay curves (within the practical limits of the detection and processing electronics); such curves relate to specific levels if the detection is sufficiently selective in wavelength (by using filters or spectrometers). Actually, as an alternative, selective excitation (by narrowband laser) would be good, but is much more expensive (find a laser that works for a wavelength band one needs!) and tedious (tuning the laser to a transition in a sample of keV-energy highly charged ions in a trap ...). The laser technique for manipulating the level populations of ions trapped in an EBIT had been tried early on at Oxford (see [35]), and it succeeded decades later at Heidelberg [36]. Given the relatively wide wavelength spacing of the spectral lines of interest for such studies, for example, the electric-dipole forbidden "corona" lines of B-like Ar ions (Ar XIV) or Al-like Fe ions (Fe XIV), they are not readily accessible by a given optical laser system. In the vacuum ultraviolet (VUV), extreme ultraviolet (EUV), or X-ray range, however, there now are tunable light sources that can fill this role. The Heidelberg device FLASH-EBIT has been transported to the synchrotron light sources PETRA (at DESY Hamburg) [37] and BESSY-II (Berlin) [38], and to the free-electron lasers FLASH (at DESY Hamburg) [39,40] and LCLS (at SLAC, Menlo Park, CA) [41]. The electron beam ion trap was used to provide a target of highly charged ions of a known element and predictable highest charge state, and the narrowband VUV/EUV/X-ray radiation was scanned across a spectral band, resulting in detectable fluorescence only when in resonance with atomic transitions (usually ground state transitions) in the target.

The electron beam energy in the EBIT limits the charge state distribution on the high side and can be varied to identify the proper charge state of the fluorescing ions. In this way even "perfect" spectral blends of lines of different charge states of the same element can be identified [38]. Such a blend affects, for example, the determination of the 3C / 3D line ratio in Fe XVII, one of the brightest line groups in astrophysical X-ray spectra. Extensive work at Livermore has long since suggested that the deviant data reported for a while from the NIST EBIT might have been suffering from a blend with another charge state, and the FLASH-EBIT experiments using photo-excitation have proved the point. A photo-excitation experiment using the FLASH-EBIT at the LCLS X-ray light source moreover corroborated the Livermore EBIT finding (in agreement with astrophysical observations and tokamak work) that there is significant disagreement between the present-day theoretical predictions of the 3C/3D line ratio in Fe XVII and experiment under even the cleanest experimental conditions realized so far [41]. By the way, Fe XVII may be the best/most researched member of the isoelectronic sequence in this case, because of its astrophysical implications, but theory (atomic structure computation) is still struggling with the 3C/3D line ratio for the whole sequence [42].

Another test of this atomic system may be expected from lifetime measurements of the 3C and 3D lines. The expected lifetimes are in the femtosecond range, but, in the first such experiment, the power of the LCLS light source turned out to be not yet sufficient [43]. By the way, the wavelength tuning of the free-electron laser can be achieved by the energy of the highly relativistic electron beam and/or by dispersive monochromators in the light path that select a narrow wavelength band out of an originally wider distribution. The X-ray laser development strives for ever more power in an ever narrower bandwidth. However, higher spectral resolution usually implies that absolute wavelength references are not easily covered in the same experiment. It would be good to have access to accurate references at many wavelengths throughout the X-ray range. Transitions in hydrogen- and helium-like ions can be computed with high accuracy. Therefore, it has been suggested to employ

such highly charged ions (with a wide selection of elements available) produced in an EBIT to serve as wavelength calibration references (via resonance fluorescence) at tunable X-ray light sources.

A bright, tunable X-ray light source such as LCLS is extremely attractive for many scientific purposes. However, it is also highly complex, expensive to build and operate, and, hence, rare. Fundamental atomic physics is a numerically small section of the prospective user community, and it takes a lot of resources to move one's own heavy and complex equipment from wherever in the world to run for a few days at one of the very few high-power X-ray light sources. Most EBIT experiments will hence stay in place at the experimenters' home laboratory and will have to do with less fancy attachments (unless the EBIT is already an attachment to a fancy facility). Even then, the actual complexity of a conceptually simple experiment may be stretching the resources of a laboratory. For example, J. D. Silver and his group at Oxford developed experiments that intended to induce (for example, by infrared laser light) transitions from long-lived levels in highly charged ions to short-lived levels that in turn would decay by X-ray emission. Thus X-ray detection would signal when the laser was on resonance and tell the accurate transition frequency between excited atomic levels. In the 1970s, the experiment began on fast ion beams, evolved through several generations of lasers and graduate students, and in the end it became a precision tool under the care of E. G. Myers (one of those former graduate students) at Tallahassee (FL, USA). The Oxford group meanwhile instigated the construction of two EBITs (one their own, one for NIST Gaithersburg) with counseling by Mort Levine, the inventor of the original Livermore EBIT. One of the experiments planned for the Oxford EBIT was again the laser interrogation of highly charged (hydrogen- and heliumlike) ions with the intention of determining atomic level separations with high accuracy. A major advantage over the previous fast-ion beam experiment was expected from the massive reduction in Doppler shift and spread, and the experiment worked [35], but the relatively small number of suitable ions in the trap combined with the available laser power density kept the true signal rate too low for the hoped-for accurate measurements. The Heidelberg EBIT group addressed the same problem via a less fundamental atomic system, Ar^{13+} with five electrons. This B-like ion offers various practical advantages. The transition of interest is the fine structure transition in the ground term, which guarantees a much higher density of target ions for the laser, and the visible laser light beam is easier to adjust (by humans) than the IR laser used at Oxford. The Heidelberg EBIT has been designed for trapping very many ions, so that enforced evaporative cooling (by using a shallow trap potential) could be employed to reduce the temperature of the trapped ion cloud and thus the Doppler spread [36]. Still, the experiment was not routine—a single transition has been studied and its wavelength determined out of the many similar ones that would be of interest in astrophysics.

In contrast to (active) laser interrogation, passive observation may not always provide the extreme accuracy achieved only by selective excitation, and it may suffer from many (mostly small) systematic errors, but it usually is much cheaper and much more flexible. For example, the hyperfine transition in the ground state ($n = 1$) of one-electron ions of heavy elements has been measured at the GSI Darmstadt by laser-induced fluorescence on a bunch of ions traveling at relativistic speed in the ESR heavy-ion storage ring [44,45]. There are results for two isotopes from this work. Concurrently, the Livermore SuperEBIT group studied five isotopes [46–48] by passive observation of a wavelength band. Some of the wavelengths were far enough from earlier expectation that a search for a laser resonance might have taken a very long time. In order to disentangle various contributions (QED, nuclear size, nuclear magnetic field distribution) to the hyperfine intervals, it seems advantageous to measure related hyperfine transitions in the $n = 2$ shell of Li-like ions. (Li-like ions are more complex, but the ground state has $n = 2$, and thus lifetime broadening of the levels and lines of interest is not an issue, in stark contrast to H-like ions.) Here, the hyperfine interval is smaller, so that the direct transition lies in the infrared. The team at GSI has tried to induce this transition for many years, and only recently a resonance has been seen, the frequency of which still needs to be located with accuracy in the rest frame of the speedy ion [49,50]. Concurrently the Livermore SuperEBIT has provided EUV/soft-x-ray spectra of $2s–2p_{3/2}$ and $2s–2p_{1/2}$ line multiplets in Bi and Pr from which the

hyperfine splitting of the $2s_{1/2}$ and the $2p_{1/2}$ electrons could be determined [51–53]. The hyperfine intervals make up only a fraction of the EUV transition energy, and, consequently, the accuracy of the determination of the hyperfine interval is lower than what ultimately the direct laser resonance experiment should be able to achieve. However, the GSI laser resonance experiment on Bi in its first decade failed to obtain a resonance signal within its wavelength range of search, while the SuperEBIT experiment on Bi demonstrated that this problem likely related to the parameter choice for the various searches, but not to a drastic failure of theory *per se*.

2.3. Atomic Lifetimes

In the earlier GSI/ESR measurements of the hyperfine transition in high-*Z* H-like ions, the time-dependent fluorescence signal was followed and a decay curve established from which the lifetime of the upper level could be derived (the initial disagreement between theory and experiment being lessened by later work on both sides). The light collection and detection design was taken over for an experiment at the TSR heavy-ion storage ring at MPI-K Heidelberg which worked without laser excitation, but utilized the leftover excitation the ions carried from the ion source and the foil (respectively the gas) stripper of the tandem accelerator that provided ions to the storage ring [54]. Over some 15 years, the lifetimes of about 70 long-lived levels in some 30 ion species were measured (for examples and references, see [55–57]). The level lifetimes in the range from 0.3 μs to a few seconds related to levels that either required a spin change (intercombination) to decay or a E1-forbidden decay. At the same time, even longer atomic lifetimes (up to minutes) in singly charged ions were studied at the CryRING heavy-ion storage ring at Stockholm, exciting and later quenching the levels of interest by laser light [58]. The CryRING machine has since been moved to GSI Darmstadt where it will serve as part of the FAIR/FLAIR facility, whereas the TSR ring is being refurbished for a new role in rare and radioactive isotope studies at the Rex/ISOLDE laboratory at CERN.

While some of the TSR measurements have achieved an excellent accuracy much better than 1%, some others were found to suffer from the efficiency of the excitation process, that is, not only the levels of interest were excited, but also some others in the same longevity class (and overlooked for a while), and the decays of the latter levels would affect the decay curves of the levels of interest [56]. Hence, it would be good to serve ions to a heavy-ion storage ring that have resulted from a less effective excitation process (that is, under low density conditions), for example, from an electron beam ion trap. However, EBITs have also been used for atomic lifetime measurements [59–62], and they have their own set of problems in the quest for atomic lifetime (high) accuracy [56]. For example, there are ions of various charge states in an EBIT, and though the residual gas density in a cryogenic EBIT is extremely low, charge exchange (CX) of ions with residual gas particles is notable (and measurable, see further discussion below). Moreover, in many ion trapping experiments one finds that the interaction of the ions in the cloud with each other matters. For precision measurements thus often ions are transported from a production trap to another one of better design for storage and interrogation. One example is the Pentatrap project for mass measurements of highly charged ions at Heidelberg [63].

The ion number after some charge state separation and energy filtering may be very small. Therefore Joseph Tan and his group at National Institute of Standards and Technology (NIST) Gaithersburg have constructed what they call a unitary Penning trap [64–66], with a small, but large-solid angle coupling lens built into the wall of the trap vessel, thus making the most of the available (visible-range) light signal. Again, this trap is being provided with ions prepared in an EBIT and then charge state selected in a transfer system feeding the external ion trap. Starting from ion trap basics, this line of development has been summarized and put into perspective by the group in a contribution to this Special Issue [67].

2.4. Microcalorimeter

In the X-ray range, conventional solid state detectors may span a large solid angle, but usually offer a very limited spectral resolution of, say, a few hundred eV, for photons in the keV range. On the other hand, X-ray spectrometers offer excellent spectral resolution, but at the cost of a small solid angle and low diffraction efficiency, resulting in a poor signal rate. Microcalorimeters, in contrast, offer coverage of a wide part of the X-ray range combined with a line width of only a few eV. There are various pathways to this goal; one practically proven design has been developed at NASA Goddard and used in its various incarnations at the Livermore EBIT laboratory [68–70]. In the latest of these instruments, the EBIT Calorimeter Spectrometer (ECS) [71], 36 individual detector pixels are mounted in an array. By choice of material and thickness, the individual pixels are optimized for low photon energy (say, 200 eV to 15 keV) or harder X-rays (up to 100 keV). The device is designed for spaceflight and operates at a temperature of some 60 mK; when an X-ray photon is absorbed, the detector element is warmed up by the converted photon energy. The temperature change affects the electric conductivity of the detector pixel, which is registered externally while the detector recovers its operating temperature via a weak link to the heat bath, an adiabatic demagnetization refrigerator (ADR) crystal. Spectra obtained with adjacent pixels can be used to veto against cosmic ray events; the cleansed and individually calibrated spectra can then be co-added to achieve a larger solid angle of detection and thus a higher total signal rate.

One of the most recent experiments with such an instrument at the Livermore EBIT has addressed the rare earth element europium ($Z = 63$) [72]. The X-ray spectrum shows very many lines from several charge states of Eu ions. In a sub-section from 1600 to 2400 eV, ten of the lines observed have been identified with n=3–4 transitions in the Na-like spectrum of the ion Eu^{52+}, helped by spectral simulations with FAC (which also indicate relative line intensities). The simultaneous observation of a variety of lines connecting to the same or related levels helps to perform cross checks in the line identification process.

2.5. Charge Exchange

In an EBIT, trapped ions may eventually be lost by several processes. Along the magnetic field lines, the ions are confined by an electrostatic potential, but the statistical velocity distribution will result in a few ions to attain a high enough velocity to overcome the electrostatic barrier. The space charge of the electron beam provides some trapping effect, but also electrons for recombination, resulting in a successive degradation of the charge state distribution. Across the magnetic field, there is no true trapping potential, but the Lorentz force deflects ions from their trajectory and thus it effectively hinders radial losses. However, the positively charged trapped ions feel a repulsive force when encountering others, and even distant collisions (without electron exchange) may change the trajectories sufficiently to result in an ion drift across the magnetic field lines and eventually to the walls of the vessel. Last but not least, and occurring even in the extremely high vacuum of cryogenic EBITs (some 10^{-11} mbar), there are collisions with neutral atoms or molecules of the residual gas. In the quest for long storage times, the charge exchange (CX)—electron transfer from the neutral gas component to the precious multiply charged ions—is a nuisance that wants to be suppressed. However, there are interesting physics aspects to consider.

Most levels in highly charged ions are short lived, and thus fluorescence ceases when the electron beam in the EBIT is stopped (the trap runs on as a Penning trap) and the trap switched to the so-called magnetic trapping mode [73]. A few levels may be longer lived, and their "delayed" decay photons then can be measured towards a lifetime determination of the excited levels (see above). If, however, the radiation seen after termination of the electron beam current is from the decay of a short-lived level, one may safely assume that this level has only then been populated, most likely by electron capture (CX) in the collision with a neutral particle. Monitoring such (mostly X-ray) events over time maps out the relative number of remaining trapped ions and thus helps to establish collisional loss rates. These are needed to correct the apparent decay rates in atomic lifetime measurements. There

is no space for vacuum gauges inside the ion trap, but the CX signal is the next best thing that even senses the environment at precisely the location where it matters.

CX has been investigated since decades in classical collision experiments, usually involving keV to MeV ion beams passing through gas targets. It has been established that the electron capture preferentially populates levels with a principal quantum number n that is on the order of the ion charge number. However, the level population may also vary in angular momentum quantum number ℓ, and the population distribution may depend on the collision energy. If levels with high values of ℓ are populated, the subsequent decays will proceed in a stepwise fashion with small changes of principal quantum number (and thus energy). If levels of low ℓ are populated, a decay towards the ground state may happen in a single step, involving the emission of an X-ray photon. Thus the photons associated with CX may have wavelengths almost anywhere in the spectral range (distributed, low-energy, "soft" spectrum) or mostly in the X-ray range (concentrated, high-energy, "hard" spectrum). The first of these is very difficult to observe, the second is much easier, but is not easily spectrally resolved.

However, one really wants instrumentation that combines sensitivity in the X-ray range with resolving power and wide spectral coverage—a case for the aforementioned microcalorimeters attached to EBITs. In highly charged ions, some of the decays with small changes in principal quantum number may already fall into the soft-x-ray range, which makes them detectable along with (but readily resolved from) the harder X-rays. EBIT is not only useful for the production of highly charged ions in this scheme, an EBIT is also able to provide the ions at kinetic energies in the keV range, in a low-density environment. EBIT measurements have already shown that the CX cross sections depend markedly on the collision energy, and they increase strongly towards lower-energy collisions. Moreover, the ℓ-level population changes significantly along the way (for examples, see [74–77]). While these CX observations challenge theory (which may be a good thing on its own after decades of calculations of a plethora of collision processes, but little prospect of generally applicable results), there is much more: per chance, X-rays have been observed from comets [78], which long since have been discussed in terms of neutral (dust) particles and singly or at most doubly charged ions in the tail but not as sources of X-rays. Presently favored seems to be an interpretation of highly charged particles of the solar wind meeting the neutral gas near the comet and there experiencing CX. The early signal samples are roughly compatible with emission after CX by highly charged ions of C, N, and O [79], but the space observations are not yet well enough resolved (and energy-calibrated?) in the soft-X-ray range. There are spacecrafts that would permit observations with higher spectral resolution (with grating or crystal spectrometers on board), but the tracking of a fast moving comet for the benefit of longer-term exposures is tricky, too. When at long last a microcalorimeter will operate in orbit, comet observations will surely be on the agenda. By the way, the EBIT work on CX shows clear changes in the spectra after CX that relate to the molecular structure of the target gases used. CX may one day not only tell about solar wind ion species and velocities, but also about the chemical composition of the dilute atmosphere of comets *in situ*.

2.6. Mass Spectrometry

In ion trapping or probing with radiofrequency (RF) electromagnetic fields, typical frequencies for protons may (for a particular trap design) be in the 100 kHz range, while dust particles have been kept in a radiofrequency ion trap at mains frequency, that is, 50 Hz (in Europe). Evidently the charge-to-mass ratio qe/m matters. In addition, higher frequencies are advantageous for higher measurement accuracy. An obvious consequence of these basic facts is that mass spectrometry of heavy isotopes should benefit from pushing the ions to high charge states q (the so-called charge breeding process), for example in an EBIT, and then transporting them to an optimum storage trap. An early version of such an apparatus was the RETRAP device at Livermore, a Penning trap that could be fed with ions produced by the EBIT-II machine. RETRAP has since moved first to GSI Darmstadt and then to Stockholm, where it it now is part of the mass spectrometry laboratory. Quite a

number of laboratories around the world are busy nowadays with accurate mass determinations for QED contributions, nuclear physics of rare isotopes, and whatever else.

The TRIUMF facility at Vancouver (BC, Canada) has a distinguished history as an isotope production and research laboratory. The laboratory has recently been augmented by an EBIT built at MPIK Heidelberg [80] for use (among other duties) as a charge breeder [81]. Isotope factories have among their problems isobars, that is, isotopes of different elements, but of similar mass number. Here, the additional tool of charge selectivity in an ion trap is expected to help distinguish between isobars. Heavy ion storage rings can be used to determine small mass differences; this is one of the reasons for (soon) moving the TSR storage ring from Heidelberg to the Rex/ISOLDE isotope production and separator facility at CERN. Since storage traps may be filled by successive bunches of ions from a source, they can also accumulate ions, which is particularly useful for the low number of particles available in isotope production. The number of ions may become high enough to try out optical detection and interrogation techniques which presently are not yet as sensitive as the RF detection techniques.

Several heavy-ion storage rings are operating for heavy-ion nuclear and atomic physics experiments. In such a storage ring, electric pick-up probes sense the fast ions passing by and build up a noise spectrum (Schottky signal) from which one can determine the accurate revolution frequency and thus derive information on the identity of individual ions stored in the vessel. If a stored ion undergoes an identity change by beta decay, the resulting change in nuclear charge and mass can be detected, and hence the dependence of such processes on the number of remaining electrons can be studied. It has been argued that certain decay channels in hot astrophysical environments (the aftermath of the Big Bang or of supernovae) may change in availability or in branching fractions, with consequences on astro-chronology, and some of the hypotheses can be tested in such ion traps.

2.7. Exotic Processes

In high-precision laser spectroscopy of trapped ions, the interactions between the ions in the trap become notable, so that often a single ion in the trap becomes the goal. In contrast, there are other experiments in which large numbers matter. More particles promise a larger signal and thus a higher statistical reliability reached in a shorter time. The Heidelberg EBIT has been explicitly designed with this advantage in mind. More ions in an EBIT also provide a stronger ion beam if the EBIT is being operated as an ion source. There may be experiments that have such a low intrinsic signal that, only by large numbers of prospective emitters, may a specific signal become detectable against the ubiquitous background signal.

A special class of experiments that are not practical in plasmas or classical light sources are those that start out from highly charged ions and imply time scales much longer than those useable in beam-foil spectroscopy. For example, at heavy-ion storage rings such as ESR at GSI Darmstadt or TSR at MPI Heidelberg, atomic and nuclear processes have been studied that depend on the charge state of the stored ion. When the number of available electrons is reduced, electron capture may no longer happen with some isotopes, or Auger decays be modified. It may be worth thinking about which processes and ions should be investigated in an EBIT instead, which is cheaper to operate, and how to arrange for optical or particle detection. Of course, the aforementioned experiments at the LCLS X-ray laser, aiming to modify the charge states of already produced highly charged ions, or to excite particular levels in these ions, are related to this class of future experiments.

For decades, nuclear physicists have hunted for an optical detection of the aftermath of nuclear decays, or for optical signatures of the reordering of atomic shells after Auger decays have ejected several electrons. Occasionally, atomic structure computations seem to indicate a promising path towards a possible signal, and in various scenarios, ion traps for selected ion species have been mentioned as important tools for the quest.

3. Conclusions

There are many more development ideas in the works than have been described here so far. A long-standing plan, for example, is the two-color interrogation of trapped ions, so that excitation by one laser pulse can be detected with better efficiency after a second laser pulse has ionized the atom, preferably by selective ionization to an excited level in the higher charge state ion because that process might be associated with a larger cross section when exploiting a resonance. The LCLS light source has offered the combination of X-rays and infrared laser pulses mostly for solid state physics but is beginning to offer pairs of X-ray pulses of different wavelengths, which would be very interesting for a target of highly charged ions prepared in an EBIT.

There certainly can be many useful investigations that vary the trapping conditions during the experiment, a line of work that started more than two decades ago with atomic lifetime measurements in the Livermore EBIT. More recently, a few such attempts have been reported [82] that played with electron beam energy and timing, involving stepwise ionization via metastable levels, and modification of the level population on the time scale of the metastable level lifetimes. Part of this work may be beneficial for atomic physics studies, and part aims at plasma dynamics.

On one hand, ion traps appear as a fairly mature technical field, now routinely applied to many physics tasks. On the other, the many improvements to devices and procedures underlying the high scientific productivity reflect the high activity level as well as a growing complexity of the experiments. From my own experience with fast ion beams, I recall the feature of delayed spectra which became viable only because our ion accelerator provided a much higher ion beam current than most of the competition, and, therefore, we could afford to let much of the prompt signal die out before addressing the leftover signal for its particular merits. A recent publication from an electron beam ion trap plays a similar scheme, evaluating spectral line intensities after a considerable waiting period, to notable scientific gain [83].

I take it that the topical range intended for this Special Issue of "Atoms" will be of continued interest and look forward to following the progress in the field.

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