

Atomic Lifetime Measurements with Ion Traps of Many Sizes

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Abstract

Atomic lifetime measurements yield information on atomic wavefunctions that supplements the data from atomic energy levels, but is more sensitive to details of the radial wavefunctions. Transition rates of intercombination (spin-changing) and electric-dipole forbidden transitions are of interest also for applications in plasma diagnostics and astrophysics. Earlier lifetime measurements of low-charge state ions in radiofrequency, electrostatic and Penning ion traps have recently been supplemented and in some cases superseded by experiments on electron-beam ion traps (EBIT) and heavy-ion storage rings. Several of these measurements of lifetimes in the range from 10 μ s to 500 ms have reached uncertainties of better than 0.5% and challenge theory. Typical features of ion trap experiments, performance problems and technical developments are reviewed, and likely routes of further progress are outlined.

1. Personal remarks and introduction

This review is not quite the type of regular review you might expect. It is not complete, and not even aiming to be so, as ion traps and atomic lifetime measurements are wide fields, and so is their region of overlap. Therefore I have elected to concentrate on the subfield and experimental techniques I am more familiar with, that is the measurement of long atomic lifetimes in ions, preferably multiply charged ones, with an emphasis on the recent developments towards precision measurements, and on lifetimes with uncertainties of less than 1%. However, “long”, “precision” and “multiply charged” will not exclude some mentioning of others.

My selection is strongly biased by my personal experiences – which also influence my views. As I will make some statements that others might see as signs of poor judgement, I would like to illustrate my way into the field, so that the reader can see on what basis I make some of my claims (The reader in search for dry facts is welcome to jump ahead to the next section.). If I mention suspicion about the validity of some data, it is not that I want to call the respective authors careless or their analysis premature – it just reflects my assumption of having experienced similar problems and being happy that either I got a suitable idea to get around the problem myself (often by realizing that somebody else had found a cure long time ago), or that in discussion with colleagues I was told of what to avoid or what to do. Anyway, ion traps and lifetime measurements began long before I took up science, and I am certain that I have not read all about them that is worth reading. Also, the complex apparatuses I used in this field (radiofrequency ion trap, heavy-ion storage ring, electron-beam ion trap) had been designed by others and for other purposes. At best I am a user of these facilities, but one with a special perspective. I am grateful to those colleagues who helped me gaining some insight and pointed out literature I had

not been aware of. I am full of awe about some people who did pioneering work so far ahead of the competition that their good work was almost forgotten by the time when the rest of the community arrived at the same physics problems and should have taken the literature knowledge into account.

One example is Wolfgang Paul who – according to his own public recollections – thought up the radiofrequency (quadrupole) (RF) ion trap but considered it to be so trivial that he gave the idea to his students to make a working device (which they did), thinking it hardly worth publishing a paper [200] about the idea. The idea, it turned out, had been around and was pursued independently by several people in different parts of the world. (In Germany the name Paul-Straubel trap has come into recent use, while Americans might call it a Langmuir trap, as it was re-invented in Langmuir’s group [264] for the English-language market, after the original publications were in German only.) For Paul himself it was just one of many of his fruitful ideas and experimental pursuits. Lots of physicists sprang from this hotbed of ideas at Bonn and became famous for their own particle trap work. With Germans finding employment at home too sparse, much of the trap activities moved to the USA during the late 1950s and early 1960s.

A second example is M.H. Prior at Berkeley who used the ion trap for some still unsurpassed basic atomic physics studies on few-electron ions [158,206,207] as well as investigations of ion dynamics (time-resolving the properties of the ion cloud in the trap) [157]. This work on fundamental aspects was done in the early 1970s, so that (I heard) he justly perceives himself as the “Granddaddy of all that”. About ten years later, Mike Prior’s student R.D. Knight moved to the Harvard-Smithsonian Center for Astrophysics (CfA) and there pursued atomic lifetime measurements on atoms of upper-atmospheric and astrophysical interest [159]. This line of experiments and their offspring will form the body of this review.

Another decade later I stumbled into the field. After more than 13 years of fast-ion beam spectroscopy, mostly doing atomic lifetime measurements by beam-foil spectroscopy, I obtained scholarships and spent two years at CfA (there would not have been any resources available to me for training years with a group in my home country, Germany, like, for example, Werth’s excellent group at Mainz). My hope was to learn about ion traps and apply them to low-charge state atomic systems which – because of their longevity – were beyond the reach of fast-ion beam techniques. However, it turned out that by that time the CfA radiofrequency ion trap and its auxiliary equipment had fallen into disrelict, to the dismay of a fresh postdoc and

a visitor (myself), both new to the field and without a local knowledgeable person to ask. Both had been told of a working apparatus and needed quite some time to realize and ascertain step by step that it was not just their dumbness that prevented the existing apparatus from working. Well, with repairs, replacements, rewiring, cleaning, suddenly available money for better vacuum, new electronics components, outside technical help from a former group member (V. H. S. Kwong, who supplied a fresh trap structure) and decisive help from the incoming next postdoc (A. G. Calamai, who at Raleigh had been trained well in trapping, by C. E. Johnson, albeit on a different type of trap), the CfA ion trap after much more than a year showed signs of life again. Then problems with the laboratory turned up and delayed further work for a while. These experiences taught me a lot about the problems and snitches of working with RF ion traps. Tony Calamai eventually managed a landmark lifetime measurement on P^+ with the revived set-up [42], which in turn elicited an impressive theoretical response (see [127]). My originally intended measurement on S^{2+} , however, took a few years longer still, and was then done by another visitor to CfA [124], now under Calamai's tutoring by "remote control".

In the somewhat depressed mood of these experiences, I attended the 1990 ICAP conference at Ann Arbor, Michigan. Complaining to German colleagues at the meeting, G. Huber (Mainz) and D. Habs (Heidelberg) bluntly asked me in return "Why don't you use a storage ring instead? It's easy, straightforward, bound to work. You doubt it? A back-of-an-envelope calculation will show you that we are right." Well, the CfA ion trap had a diameter of about 3 cm, whereas the Heidelberg storage ring (the actual construction work of which had derailed a slow-beam experiment of mine a few years earlier) has a circumference of about 55 m and costs correspondingly more to build and run – that idea had simply been beyond my horizon. The overnight estimate indicated that the experiment might be feasible, indeed, though barely, in the face of photomultiplier noise, assuming ion beam parameters as presented at the conference. After my return to Germany, and two funding cycles of the proper German research ministry later, the experiment succeeded, at long last – and at the first try [78]. It worked better than feared (in signal rate) by about two orders of magnitude, and it yielded an atomic lifetime with a precision (0.13%) quite close to the best ever achieved at the time on any atomic system. Last, but not least, it improved on a measurement from the CfA ion trap by a factor of 50 in precision and also suggested the presence of unrecognized notable systematic errors in that earlier work (a shift by 5 standard deviations).

When working at the Harvard–Smithsonian Center for Astrophysics, I had enquired – in vain – about the final outcome of their important RF ion trap experiment on C^{2+} , preliminary results of which had been shown at a conference years before [228]. For lack of anything else, these data were then accepted into a semi-empirical analysis of the iso-electronic trend [64]. Years later again, a formal result was presented by the principal investigator [164] that was so different from the first one that not even the error bars overlapped. However, instead of being cautioned by these diverse experimental results from the same authors (who did not offer any explanation of this discrepancy), two theor-

etical studies appeared that happily claimed agreement with the new data. Other theorists tried to work systematically on this fundamental system and then decidedly disagreed with the experimental numbers [100,106,269]. In fact, their results, backed up by convergence studies and error estimates (smaller than the last experimental errors), lay in the gap between the error bars of the two published results. When, soon after, experimental data from a heavy-ion storage ring experiment [78] perfectly corroborated these latter calculations, they spurred further theoretical improvements (now that the basic discrepancies were settled) [107] until all of the better experimental data, for the singlet (up to $Z = 36$) and triplet (up to $Z = 54$) resonance levels, could be represented rather well by a single calculation [143]. At a conference I met Leonid Vainshtein, co-author of one of the two theoretical studies [50,210] that (after the measurement) had agreed with the second experimental result from the RF ion trap. I jokingly reminded him of my earlier warnings and asked him why he nevertheless had believed in the numbers from his atomic structure code. The answer was: "Why, at the time experiment was there!" So much for the predictive power of (some) theory.

Theory does have predictive power, nevertheless: Some of the same theoreticians who did the C^{2+} system right – in the face of contradictory experimental evidence – also did calculations precise and accurate enough to cast doubt on the earlier, very precise ion-beam – laser lifetime experiments on the resonance levels in Li and Na, and they were vindicated perfectly by the recently improved new measurements by various groups and very diverse techniques (see [252]). Also, in order to disperse the reader's suspicion that experimenters ever look at theory before trusting their own results (unfortunately, sometimes there are no theory data to compare with ...), I might say that I enjoy having diverse predictions. Theoretical guidance to the right ball park is, indeed, very useful, but from then on a slight disagreement is a helpful incentive to make certain about the experimental precision (getting a small uncertainty) and accuracy (making certain about the mean value given). However, the nicest case is to select among a range of predictions the one that turned out best and to notify and praise the colleague who got it right. As certain colleagues have turned out to be right more often than others, one is tempted to assign the predictive power not to "theory", but to certain theorists and their particularly meticulous work. Unfortunately, some of them are so well aware of their possibilities that they refuse to tackle some of the problems the experimenters want numbers on, as being "too complex for my code".

The beautifully simple and reliable heavy-ion storage ring technique has since produced a few more neat numbers, and as the systems of primary interest are not so many, there is more overlap with the earlier CfA work. While in most cases the error bars can be reduced, some of the early work is being corroborated by the better measurements, and some is not. My impression is that – perhaps by virtue of the particular experiments, perhaps by other factors – some authors' studies appear to more likely withstand later scrutiny than others. I am, for example, impressed by the fact that so far I see no contradiction of the heavy-ion storage ring results to the data obtained at CfA by B. Carol Johnson. For a number of other data the situation is

different, and discussions with people involved give me the feeling that some elder colleagues are right. They say that for a preliminary assessment of the quality of data, be they theoretical or experimental, it is often better not to look at the sales talk in the abstract (on the fancy computer codes used) but to look for who did the work. In a similar vein (and having the elementary particle physicists' Charm, Beauty and Truth in mind), I am inclined to add "Arrogance" as an ill-understood, but almost certainly significant ingredient to the list of often overlooked systematic errors ("Positive Thinkers" may use "Chuzpah" instead.). I know of several cases (not all related to ion traps) in which I would blame this as the real reason why published data later turned out to be wrong, sometimes by a high multiple of the stated overall error. One author introduced as a "small, but significant change" of his own earlier data what, after two iterations, amounted to a correction by six of the originally quoted standard deviations, plus enlarged error bars. Unfortunately, some of the (in hindsight) faulty data have made it into data collections and compilations. With the funding for compilations declining and many of the arduous compilers of spectroscopic data nearing pension age, the faulty data will be fossilized in the libraries. At several occasions I learned how many people rather consult an outdated compilation than the more recent original literature. Maybe electronic data bases and search facilities will help a bit against this intellectual sloppiness.

Back to more regular science talk. In my meandering through atomic physics, I have over the years measured a number of atomic lifetimes from a few picoseconds up to, say, 30 ns using fast ion beams [14,15,150,231,238], had one student measure a 200 ns lifetime on a slow ion beam of heliumlike Ar¹⁶⁺ ions [134], had another one try the external trapping of recoil ions with insufficient means, aiming at microsecond lifetimes [125], and jumped to the millisecond range with RF ion traps and the heavy-ion storage ring (with lifetimes of almost up to a second). Because of technicalities explained below, the latter can store intense beams of highly charged ions, but that process often takes longer than the ions of interest will persist in excited levels. It is therefore desirable to have a device that produces highly charged ions more easily and directly, and that permits their fairly unperturbed study in the time range of, say, up to 1 s. Such a device is the electron-beam ion trap (EBIT) that was developed as an off-spring of the electron-beam ion source EBIS. This gives me another chance to present an example of the none-too-straight ways of scientific progress.

The aforementioned experiment on a recoil ion beam of Ar¹⁶⁺ [134,135] aimed at nailing down a hypothesis why a fast-ion beam experiment at Berkeley [184], much earlier, had resulted in a lifetime number that was massively (20%) different from prediction but had been presented with high confidence expressed in fairly small error bars. At the HILAC accelerator, which was needed to reach the high charge state by energetic ion-foil interaction, those authors had faced a decay length (ion lifetime τ times ion velocity v) of almost 8 m, of which they could measure over only a fraction of less than a quarter, and the experiment possibly also suffered from an incomplete suppression of core-excited three-electron ions [171]. In contrast, the slow ion beam of the later recoil ion experiment reduced the decay length to 6 cm, which was quite manageable for our 10 cm long

low-budget, little-experience, position-sensitive X-ray detector. (The basic idea to this detector and its application I had as postdoc at Oxford – only to find that Richard Mowat had already independently published the idea of a similar device [195]. Precursors to this idea of recording decay curves with a fixed, but position-sensitive, detector were the photographs of radiating ion beams taken in the early beam-foil days, and a later perfection of the same idea can be seen in Kay's design of a telecentric imaging system coupled to a charge-coupled device (CCD) camera [151].)

In parallel to the recoil-ion beam set-up for the parasitic ion beam at the stripper hall at GSI Darmstadt (and paid from the same little grant that paid a single graduate student) we prepared a system to work on a decelerated ion beam at the Heidelberg facility, where a post-accelerator could alternatively be tuned to slow down fast ions (which were needed in a first step to reach the proper high charge state), down to a fraction of their former energy. Our partner there was Reinhold Schuch. By the time we had our experimental chamber built and wanted to do the experiment in earnest, the construction of the heavy-ion storage ring TSR blocked the intended location, and Schuch moved on to Oak Ridge and to their heavy-ion storage ring project that was canceled soon after. Schuch then got a position at Stockholm where another storage ring (CRYRING) was being planned. For this ring, an ion source of the EBIS type was being set up and tested. This source was deemed to deliver enough slow ions of the right charge state for a possible re-run of the Ar experiment, and Schuch invited me to try an improved version of that. He readily constructed a new position-sensitive X-ray detector that promised a performance much superior in efficiency to our poor first version. However, the cryogenic system of the ion source had recurrent problems after the Swedish summer break, and nothing came of the Ar project then. I moved to the USA (the above stint at CfA) and from there could not provide any help or money for setting up a beam line, which Schuch then did on his own. However, in his pilot trials, there was not enough ion current passing through the line to the detection site, and the experiment has been dormant since. As will be discussed later, it still would be very interesting to do this experiment and realize its potential precision, even 10 years later. However, while at the time a precision of 6% was state-of-the-art, theoretical progress now calls for testing at the level of 0.5% and better.

The Stockholm cryogenic EBIS (CRYEBIS) was not the only EBIS that had intensity problems [79], and Mort Levine at Berkeley realized that such problems might be less pronounced with a shorter device [168]. Not taking the ions out of the source gave the kid a new name – it became the electron-beam ion trap – which nowadays is not only used as an ion trap, but also as an ion source. Two EBITs were built at Livermore, then two more at Oxford (one of them for NIST Gaithersburg), another one meanwhile is being up and running in Japan, and yet another one in Berlin. Frankfurt does EBIS/EBIT design and performance studies, Freiburg is setting up an EBIT for an ion source, and other laboratories have plans to get one, too. I had occasional projects in collaboration with the NIST spectroscopy group, and at one occasion I was asked by John Gillaspay whether I had interest in joining up with their EBIT group as well. My interest was in atomic lifetimes which, I

learned, had also been brought up by Dave Church and his graduate student who was then working at NIST. (Church has done many things in the ion trap business, including an extensive review [53].)

Collaborating by occasional NATO-sponsored visits and mostly by e-mail, we obtained decay curves on some forbidden transitions in highly charged ions. At that time, Peter Beiersdorfer at Livermore had already performed demonstration lifetime measurements on just the same transition in heliumlike ions as the above Ar^{16+} experiment covered, but on several lighter ions, modulating the electron beam energy in EBIT around the excitation threshold. By the time of the NIST lifetime measurements, in which the electron beam current was gradually reduced or even switched off completely, he had been using the much more extensive Livermore diagnostics systems to ascertain that this mode, “magnetic trapping” without the electron beam, was indeed suitable and permitted highly-charged ion storage for many seconds [17,19]. He also checked for systematic errors by comparison with the first heavy-ion storage ring results [215]. Subsequently, Beiersdorfer and his group improved on the precision of their pilot measurements by considerable factors [57,242].

Suddenly a new situation emerged from the puzzle pieces: While some groups continued to work on the perfection of traditional ion trap measurements and over time achieved some (few) data with uncertainties as low as 5% for singly charged ions, here was a new set of experiments, using heavy-ion storage rings and EBITs or trapping ion beams electrostatically, to achieve the same or better for multiply or even highly charged ions (up to $q = 32+$ so far [223]), and with the potential for quite a few more interesting studies of different points of interest. I am happy to be involved with several of these efforts which bring additional physical and technical insight by their interaction, and I hope to convey some of this fascination in the subsequent, more traditionally styled, parts of this report.

2. Scientific introduction

2.1. Basic atomic physics outline

The study of atoms has many aspects. Leaving aside the response of atoms to outside influences (collisions, external fields), we pretend that there is the atom *per se* that ought to be understood from its emission spectrum, that is from the light emitted after whatever cause has excited the atom. The fact that the light spectrum (in emission and absorption) shows structure points to the level structure of the atom. The next observation, that not all level differences appear as lines in the spectrum, and that existing lines have characteristically different relative intensities, points towards selection rules which reflect symmetries of the atom and its interaction with the radiation field [80].

There are transitions (between levels k and i) that take place with a high transition probability A_{ki} , and others that are less probable by orders of magnitude. An inspection of the symmetries involved reveals that the most probable transitions of the atom correspond to electric dipole (E1) transitions which connect levels of opposite parity (Fig. 1). A series expansion of the radiation field has magnetic dipole (M1) and electric quadrupole (E2) components next (and so

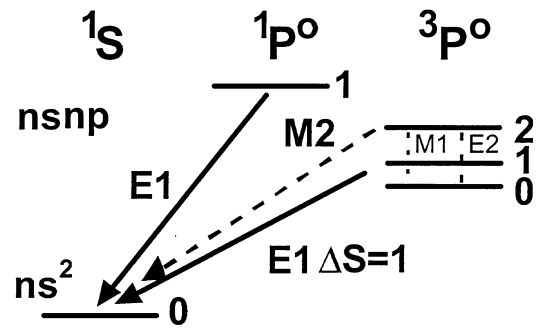


Fig. 1. Level scheme of Be-like ions, indicating various transition types.

on, M2, E3 ...), but they usually are of much lower transition probability (and connect to other levels, of the same parity), so that they do not matter as long as an E1 transition is allowed. However, sometimes this decay path is blocked for an atom, and there may only be lower levels of the same parity, for example the other fine structure levels of the same term (level multiplet). Also, the multipole transitions scale differently with the nuclear charge along an isoelectronic sequence (ions of different elements, but with a constant number of electrons). Thus in highly charged ions the higher-multipole order, electric-dipole “forbidden” transitions are relatively more prominent.

E1 transitions are your ordinary type, and their calculation is closest to the hydrogen system (and hydrogenlike ions) that theory can do well. Typical level lifetimes (the inverse of the sum of all decay probabilities from a given level) are in the nanosecond range for neutral atoms and become too small (sub-picosecond range) to measure with precision for only moderately highly charged ions (E1 $\Delta n \neq 0$ transition rates scaling with Z^4 , with the principal quantum number n). However, transitions that require a spin change (E1 $\Delta S = 1$, *intercombination* transitions with a change of spin S connecting singlet and triplet, or doublet and quartet, term systems) or that are E1-forbidden, have a very small probability in neutral atoms and may become measurable only in multiply charged ions because of the steep increase of their transition probabilities with the nuclear charge Z . These increases have long ago been systematized by Hylleraas [137] and depend on two factors. One is a dependence on the transition energy (which itself has a characteristic trend, like the Z^2 dependence of levels of different n in Bohr’s model of the hydrogen atom which describes atomic shell separations, or the linear dependence on Z of energy differences within a given shell of a multi-electron system ($\Delta n = 0$)). The other is the “physics” part, be it expressed as line strength, oscillator strength, multiplet mixing parameter, hyperfine interaction or the like. For a more detailed introduction, see, e.g., [123,94].

There are Z -dependences like Z^{10} (for the E1 intercombination ($\Delta S = 1$) transition probability with a change of principal quantum number n , or the M1 transition between the lowest levels in the He-like ions) or Z^8 (for the M2 transition with $\Delta n \neq 0$), and so on. Important for the present discussion are just a few general points:

- A decay mode of interest may be accessible to study only in a few ions, where it is not dominated by some other decay or competing collision processes.

– A transition of interest may be accessible to a given instrumental technique only in a particular range of ion charge states, because of its transition energy (wavelength range) or lifetime. For example, fine structure intervals scale with Z^4 , and thus the wavelengths of the associated forbidden transitions vary drastically along the isoelectronic sequence. This is a problem for theory, too, which is still struggling with *ab initio* predictions of such fine structure intervals in complex ions to better than, say, 10%.

Intercombination and forbidden transitions in low-charge state ions are of great interest in astrophysics, because their observation reaches larger optical depths than the observation of allowed transitions. In higher charge state ions, intercombination and forbidden transitions are of value for the diagnostics of solar and terrestrial plasmas [84,91–93,108,174]. Forbidden transitions in singly charged ions are the basis for many cases studied in pursuit of frequency standards, because the low transition probabilities result in small level and line widths. Forbidden transitions in neutral atoms are being studied in the context of parity-violation measurements, because the relatively large effect on something that is normally “not happening” (the forbidden transition) is sometimes better observed than a minute change to something that regularly takes place (like fully allowed transitions). However, only very recently any precise measurements (accuracies in the single-percent range or better) have been achieved on intercombination or forbidden transition rates in any ion. Because of the need to contain the ions under study for long enough periods, all these experiments involved ion traps, of which there are many types and working principles. Such atomic lifetime experiments in diverse ion traps are the subject of this review.

Although for some practical purposes and for the atomic clock or parity violation measurements possibly only the order of magnitude of the long atomic level lifetime may be needed, not the actual lifetime values, the quest for a detailed understanding of atomic structure is certainly helped by independent checks on the quality of the wavefunctions used. The variational principle identified by Ritz only applies to energy levels (any imperfect wavefunction will yield energy levels higher than the true ones). E1-transition probabilities, in contrast, depend differently on the wavefunctions, in particular on the radial wavefunctions. The latter need not be orthogonal to each other and are nowadays optimized separately for initial and final states of a given transition [102]. In the absence of a guiding principle from theory, only experiment can provide decisive tests of atomic lifetime calculations.

Let me make one more excursion to show the general picture. The challenge for theory is very different in neutral atoms and multiply charged ions. In atoms, there is a weak central field, but plenty of electron-electron interaction (correlation) in comparison. In highly charged ions, the central field dominates, and relativistic effects are important or even overwhelming [153]. For example, theory has problems with the predictions of intercombination transition rates in low- Z systems, because there the multiplet mixing is very weak (the Breit term is rather small or, in a relativistic picture, cancellation of the wavefunctions that yield the

wanted transition probability is almost complete [143,269]). The present trend in computations of atomic structure, even for near neutral systems that are dominated by electron–electron correlation, are relativistic calculations using extremely large basis sets. Clearly progress is being made in treating such complex systems, and it is helped by cheap computing power.

It is generally found that calculations converge much better with the strong central field case. Thus it is expected that theory does better for multiply charged ions and ought to be tested there first. In the long run, however, the goal is the better understanding of our environment, which is composed almost exclusively of neutral matter. Neutral atoms can be studied with extreme precision by laser light, and minute effects like parity violation can be investigated in heavy (high- Z , many-electron) neutral atoms and tell us about fundamental aspects of the building blocks of the universe. Theory does quite well for one- and two-electron systems of all charges, and theory is ahead of experiment in the precision determination of quantum electrodynamical effects in, say, hydrogen-like U^{91+} ions. Theory also pointed the way for the transition probabilities of the resonance lines in light alkali atoms, in which slight mis-estimates of systematic errors in earlier fast-beam laser experiments were only recently overcome by a number of different experiments, now in satisfying agreement with theory at the 0.1% level. Some of the same experiments, however, also cover heavy alkali atoms, and find the same type of theory that does well for light atoms to be wrong for heavy ones by up to 20% [252]. We will discuss similar cases (at a lower level of precision) with ions.

What I would like the reader to keep in mind is the need for tests of very different systems and in very different charge states. Many of the existing data and calculations will be good enough for the applications mentioned below – after all, the interpretation of the intensity of a spectral line that is barely seen at all in the spectrum of a remote astrophysical object usually does not require transition rate data that are more precise than, say, 10%, nor does the diagnostics of a temporally varying fusion test plasma. However, with the advent of precision measurements, the hunt is on for tools that help our fundamental understanding of atomic structure, and here precision lifetime measurements can play a very special role.

3. Trapping and detection

Ion traps of various designs have become standard tools for many physics experiments and applications, from mass spectrometry to the development of ultra-stable frequency standards. Material of an enormous price range has been trapped, ranging from “dirt” (in the quest for the scattering signatures of interplanetary dust) to antiprotons. At the same time the use even of simple ion traps reaches from applications like rest gas analysis and the composition of the solar wind (both using quadrupole mass filters that are close relatives of radiofrequency ion traps) or the development of ever more stable atomic clocks to fundamental physics like the mass spectrometry of normal and antimatter [55,72,73,109,110,142,249]. The basic working principles of the conventional electrostatic (Kingdon [154]), magnetic (Penning) [204] and radiofrequency (Paul [200]) traps have

been outlined elsewhere [70,99,219,258,259,264], as have those of electron-beam ion traps [116,168] and ion storage rings [196], and need not be repeated here. Instead, this review aims at presenting one particular application of the various ion traps to one particular atomic physics problem, the measurement of the mean lives of excited atomic levels in stored ions. This is regularly done by monitoring the spontaneous light emission from the stored ions, although other probing schemes (like laser-induced fluorescence) are known as well [140,141,157,167,212].

This review attempts to show how experimental progress of atomic lifetime measurements that use various types of ion traps, from millimeter size [25] to that of an indoor sports arena, is being made to match the theoretical quest. As the reader will see, similar physics problems have been tackled by using different types of traps, and different physics topics have been pursued with a given trap. Therefore it makes little sense to systematize the efforts by the trap type, the atomic system and its complexity, by transition type, by wavelength, by lifetime or charge state range, except in order to mark some entry points into this highly interconnected fabric. Many details can be found in earlier collections, reviews and books [27,41,53,54,115,116] or tracked from there. Here we concentrate only on a section of the panorama and discuss the general aspects but briefly. The complexity of the field will nevertheless lead to some information being repeated or referred to under different aspects even within this review.

3.1. *Ion production and storage*

The question arises of how to put ions into a trap. Without dissipative forces (collisions with a rest gas, or optical molasses) that may reduce the total energy of some particles, ions sent into a trap will simply pass through and leave it again. This leaves two options. Either the trap is kept running while the ions are produced inside the trap volume, or the trap is open during injection of charged particles and then suddenly closed. The first option usually proceeds by electron-beam bombardment of either a gas or laser-ablated [145] or ohmically produced vapour [56] in order to produce low-charge state ions, or by an energetic ion beam or intense synchrotron radiation to produce slow, highly charged ions [51,52,208]. Any such production from the ambient gas carries the penalty of the ambient gas still being around when it is no longer needed nor wanted, that is when one wants to have clean storage conditions. Only under ultrahigh vacuum (UHV) one can expect storage times that exceed a few seconds, but it is difficult to produce sufficiently many ions from such a dilute rest gas so that an optical signal may be seen. Alternatively, the trap is briefly switched off while an ion species is introduced, which is then captured by raising, say, electrostatic potential walls around the sample [54,67,109,110,125,219,270]. Such external ion production and subsequent capture and trapping, or production in one trap and transfer to another one at lower base pressure, have brought about major reductions of the associated errors.

Producing an ion cloud inside a trap (or moving there a sample of unmatched geometry) brings about the problem of ion cloud dynamics [157]: a freshly produced sample of charged particles needs to find a new dynamic equilibrium. While the ion cloud settles and thermalizes,

there is additional ion loss from the ion cloud, and controlled measurement conditions can be expected only later. This makes lifetime determinations below about 1 ms difficult (although there are data down to about 0.05 ms [124,144]). Laser-excitation of ions in a settled-down ion cloud might reach much shorter lifetimes eventually, as it has with molecular ions [176,177]. However, there are not many transitions in excited, in particular multiply charged, ions that could be forced selectively by laser radiation, because there are few suitable short-wavelength lasers. On the other hand, some extreme cases of much longer lifetimes have been addressed in singly charged ions (see the discussion about atomic clock ions below).

3.2. *Storage time constant*

The optical signal from an ion cloud that contains excited particles decreases with time, because atomic de-excitation takes place (that is the process of interest) and because other processes (charge exchange, large-deflection angle collisions) take place that reduce the number of ions (of the proper type) in the stored sample. The rate of these loss processes needs to be established in order to correct the apparent optical decay rate and arrive at the value of the optical decay rate that would correspond to the absence of perturbations. To this aim, one may, for example, eject the ions at different times after starting the trap cycle, assuming that the detected ion signal is proportional to the number of ions that survived in the trap up to the time of ejection. There are traps with a preferred axis along which particles can exit so that the majority of the ions can be seen by a single detector. There are other traps without such ejection along an axis of symmetry, and then the ejected ions spread out in a wider solid angle. This reduces the statistical significance of the ion signal, as only a small fraction of the total number reaches the detector. Moreover, any possible geometrical intricacies of the trapped ion cloud are neglected. However, a time-of-flight analysis of the ejecta may yield additional information on the charge state composition (and possibly the momentum distribution) of the formerly trapped particles.

In some trap types, non-destructive measurements are possible, for example by analyzing the Fourier transform (FT) of the spectrum of ion cyclotron resonance (ICR) frequencies (FTICR). With many multicharged ions in the trap, however, this technique does not necessarily yield information on specific charge states. Whatever the technique, it tells about charge states at best, but not on the fraction of excited ions, and in particular not of the excitation level of interest.

Ion traps serve to store ions for extended observation times, and this requires conditions that are as clean as possible. In particular the rest gas pressure has to be very low, so that stored ions do not get lost too rapidly by charge-changing collisions with the neutral ambient gas. The cross sections of such collisions increase rapidly with the ionic charge, making it more difficult to store multiply charged ions than singly charged species. Collision cross sections (for excitation, ionization or electron capture) generally decrease – beyond a certain maximum value – with increasing collision energy. While this would seem favourable, more energetic particles require deeper traps for storage, but in a given collision they also reach more

highly excited levels or even lead to ionization. Both, electron capture and loss, need to be considered for optimum working conditions.

As noted above, the ion loss rate often is obtained from measuring how many ions are left in the trap after a given period. However, it is well established that excited ions can have much larger collision cross sections than ground state ions [133]. Thus any such measurement is systematically biased: It measures the survival of ions, a fraction that is increasingly dominated not by the excited ions of interest, but by the ground state ions that are not optically observed. In fact, excited ions are in the minority right from the beginning (it would be very interesting to find a technique that excited the majority of ions in a sample). Consequently, even a systematic study of ion survival times does not say much about the small fraction of ions in long-lived excited states. This problem is illustrated by decay curves recorded with an ionization detector at a heavy-ion storage ring [77]: An almost constant signal (over a storage period of 200 ms) from ground state ions carries a small (few-percent amplitude) decay curve on top that is attributed to an excited level with a lifetime of order 1 ms.

In principle, this problem is known since decades. Ion accelerators, in particular of the tandem design, often incorporate thin foils or internal gas targets (strippers) in order to change the charge of the ions and thus achieve better acceleration or beam guidance. The same ion-foil or ion-gas interaction is the basis of beam-foil spectroscopy [14,15,150,238], and the interaction of the ions with the rest gas determines the survival (storage) time of ion beams in ion storage rings. Depending on the balance of electron loss (ionization) and electron capture (recombination), both with different energy dependences, an ion beam will develop towards an equilibrium charge state distribution. For low energies (eV-range as in conventional ion traps) the recombination processes from collisions with the residual gas (charge transfer) will regularly dominate, whereas for high-energy (multi-MeV) ion beams the rule of thumb calls for maximum charge state (bare ions) to reach maximum storage time. There are various formulae to estimate the mean and the width of such equilibrium charge state distributions, and many of them apparently have been derived from the pioneering description given by Dmitriev and Nikolaev (see [74] and references therein). The original formula contains, among other parameters, the ionization energies of the various charge states. This clue can be exploited to estimate the ratio of collision cross sections for ground state (full binding energy) and excited state ions (the less tightly bound active electron implies that less energy is needed to achieve ionization from there). This effect may be considerable in low-charge state ions, but is less important in highly charged ions, as the ionization energy scales with Z^2 , whereas the energies of typical long-lived and low-lying levels scale only linearly with Z (for s-p-d excitations). Furthermore, at high energies, any of the electrons may be involved in a collision, not just the most weakly bound. Moreover, the Dmitriev approximation is intended for energetic collisions (beyond the Bohr velocity) and thus not applicable to “thermal” collisions in conventional ion traps where – as mentioned before – excited state cross sections may differ by order of magnitude.

In short, there is no simple recipe to establish the collisional loss rates specifically for an atomic state of

interest in a lifetime measurement. The traditional method calls for measurements at various pressures and an extrapolation of the apparent decay rate towards zero pressure. The signal to be evaluated for decay curves regularly is better with more particles in the trap (higher pressure, say in the 10^{-7} mbar range), and that implies in turn that the data at the lowest pressures (often not better than 10^{-8} mbar) regularly bear the largest error bars – and thus cause a notable uncertainty of the extrapolated trend. Clearly any method that permits measurements at lower vacuum pressures (and thus does away with the need for extrapolation) is to be preferred.

Correcting measured (apparent) lifetimes for the ion storage time always results in a longer “true” lifetime value. An insufficient correction, like one that is based on (experimentally accessible) ground state ions instead of the generally shorter-lived excited ion population, will fall short of the goal and yield a lifetime value that is on the short side of the true atomic lifetime. There are a few cases for which comparison data are available from ECR ion source plus Kingdon trap, EBIT and the heavy ion storage ring (the device that has the longest ion storage times and hence the least need for correction). In the sub-ms lifetime range, storage time corrections are very small, and results from different trap types agree with the same trend [242]. In the case of multi-ms lifetimes, however, the storage ring results often are the longest, suggesting insufficient storage time corrections for the others [243].

3.3. Detection

In a classical ion trap it is difficult to probe the state of stored ions except by active interrogation with (laser) light or by passive photon spectroscopy. Both will be discussed below. With the tube trap [67,270] discussed later and with heavy-ion storage rings, however, particle detection, after state-sensitive processes that changed the charge state of the stored ion species, is possible. Particle detection has the advantage of a high detection efficiency – for energetic, fast ions the efficiency is practically 100%. However, this technique is so specific for the storage ring that the discussion will take place there (Section 3.4.3).

Laser spectroscopy has found many ways to detect small numbers of atoms, be it for trace analysis, toxicology or environmental monitoring. Some of the most sensitive techniques involve ionization processes, because the detection of the ensuing charged particles can be effected with high efficiency. Applied to particle traps, however, this would be a destructive analysis tool. Under most circumstances it would seem preferable to obtain the wanted information non-destructively, so that the experiment can continue and thus the same ions be probed again and again. The obvious choice is laser light, as there may be many photons (promising some interaction even as the cross sections may be small) of well-defined energy (promising state-selective probing). Unfortunately, there are a number of problems with trapped ions that limit such an approach to just a few cases. For example, in ions the energy splittings between terms generally are larger than in atoms. The resonance lines (ground state to first excited levels) are often in the ultraviolet (UV), maybe even in the vacuum-uv (VUV), where suitable lasers (powerful and tunable) are

rare. This may change with the progress in VUV lasers, but it certainly adds complications.

Detection may have to resort to indirect means. In some ions studied for applications in atomic clocks, for example, the laser interaction is not on the clock transition, which has to be of an extremely low transition probability (which corresponds to very little level broadening). Instead, a laser probes the population of the clock level (or related levels) by exciting the ion from there to other, higher-lying levels. The fluorescence from the decay of the latter levels tells if that absorption was possible, that is, whether there was any notable population of the level of interest. Probing the level population thus can serve as a tool for a lifetime measurement on a level without observing the spontaneous decay of the latter directly. In this way one of the clock levels ($\text{Yb}^+ \text{}^2\text{F}_{7/2}$) has long since been demonstrated to have an extremely long lifetime [167,212], of 10.1^{+7}_{-4} years.

Laser excitation of particles in a settled ion cloud can also be used with molecular ions, in order to probe specific levels after internal vibrations have decayed away [176,177]. Similarly, a heavy-ion storage ring has been used to store a beam of molecular ions and to extract it only after much of the internal excitation of the molecules had been dissipated by spontaneous radiation. Then the beam of near-ground state molecular ions was extracted and used for Coulomb explosion imaging studies [9,101]. As the structure of the molecules of interest depends on the inner excitation, the observed variation of the Coulomb explosion pattern with the elapsed storage time reflects (a multitude of different) molecular level lifetimes. It may well be possible in future to study the lifetimes of peculiar molecular ion states, if a sufficient fraction of the molecular ions assumes a specific (geometric?) configuration that lives long enough to survive the de-excitation of other states while the ion beam is stored, and then yields a Coulomb explosion pattern that can be discriminated from the regular (near-) ground state one.

In most ions, laser excitation from the ground state of ions is difficult [120]. On the good side, there are some excited levels in certain ions that are so long-lived that they may have survived from the time the trap was loaded with ions, and laser probing may start from those and then work in a more convenient wavelength range, as the separations of higher (principal quantum number) n electronic shells are usually much smaller than that first step from the ground state.

Atoms in typical samples (gas cell or atomic beam from an oven) have energies well below 1 eV, and many orders of magnitude less than that if any cooling can be effected. The Doppler spread thus is moderate, and in many cases Doppler-cancellation techniques can be applied and narrow spectral lines be observed or exploited. In contrast, the production of ions which are to be stored in a trap is often rather violent, and the ions are then stored at energies of several or many eV which entail considerable Doppler widths of emitted or sensed radiation. Much of the sensitivity and selectivity of laser-based techniques therefore is unavailable with trapped ions, unless significant cooling is being achieved first. Consequently an important feature of suitable clock atoms is their capability of being laser-cooled. Then the interactions between the ions become so important that ultimately only single trapped ions are wanted for precision

studies of their internal structure or for the reproducibility needed in frequency standards. Cooling by whatever technique, however, costs time. There are many atomic ions that have lifetimes long enough to need trapping for the lifetime measurement, but too short to merit cooling, and with internal structures that make laser techniques inappropriate or unavailable, at least for the time being. Most of the cases discussed below are in this category.

With ions that do not have energy intervals that are compatible with available lasers, the problem shifts to the detection system: Of course, detectors for all wavelength ranges are available, but the detection of weak signals (from the small fraction of excited ions, in the range a few to a few tens of percent, among the not so large number of trapped ions) depends on the detection process and its efficiency. For light detection it involves the solid angle of detection, the coupling of the light beam to the detector and the quantum efficiency of the detector as well as the intrinsic detector dark rate. Much of this is closely correlated to the trap design and depends specifically on the various trap types discussed below. However, a few points are common to various traps.

Photon detection, be it of fluorescence after laser excitation or of spontaneous radiation after whatever collisional excitation, profits from a large solid angle of detection in order to capture as much of the available light as possible. However, ion trap geometries impose certain limits. The statistical significance of the detector signal depends on both the absolute number of detected photons and on the signal-to-background rate. For typical photomultipliers, the dark rate is a function of the cathode area size. This makes a small cathode desirable, but it may be difficult to image an extended ion cloud onto it with high efficiency. While small ion traps have been built for single ions or μm -size ion clouds, other traps (and ion clouds) have sizes in the cm-range, while storage rings have circumferences of 20 to 100 m. In the latter cases, the extended light source is more easily accommodated by a larger cathode – at the cost of some dark rate. In the UV, the index of refraction of many materials approaches infinity – which would be good for short-focal lens lenses, if it would not imply strong absorption. UV-grade materials, however, have a rather low index of refraction in their range of applicability. For large-solid angle lenses this necessitates small radii of curvature (thick lenses), which introduce imaging errors, vignetting (limited solid angle because of internal total reflexion) and absorption; multi-element lens systems suffer from light loss at the many surfaces. Window materials for ultrahigh vacuum (UHV) vessels are of limited optical quality (causing imaging and reflexion losses), window flanges (glass-metal seals take space!) are often bulky and reduce the solid angle of observation. Various compromises are usually needed. For example, at an ion storage ring (or a tube-like ion trap) there are the options of sideways observation through a window close to the ion beam or ion cloud, respectively, or along the ion path (from a bend of the storage ring at any of the dipole magnets). In the former case, a section of the ion beam (cloud) of about the size of the window (say, 5 cm diameter) can be observed. This viewing region can be expanded by using an assembly of several detectors (and windows) in parallel [155], or by a system of light guides (for visible light) that serves a single detector

and thus keeps the dark rate low [257]. In the latter case, observing the ion cloud or ion beam along the tube, a much longer section is in view. However, most of this longer section is farther away from the window and thus subtends a smaller solid angle. After convolving light source size and solid angle, one of the options may turn out as preferable in a particular case – but there is no universally advantageous geometry. With fast ion beams, Doppler shifts and spreads add complications, advantages and disadvantages to a given design.

With EBIT, the ion cloud is produced and confined in a volume given by a tightly collimated electron beam (typical diameter 70 μm) [247], while slots in the innermost shroud as well as windows and flanges limit the viewing zone to typically 2 cm in length. In order to maximize the light use, it is advantageous to use this emission zone as the “entrance slit” of a spectrometer, without need for further collimation and imaging. EBIT is very good to use with a multichannel (position sensitive) detector like a charge coupled device (CCD) that is positioned in the place of the traditional exit slit. With present pixel sizes of, say, 25 μm , this is quite a good match. However, finer spectral resolution (with future finer-grain CCDs) can then not be achieved by reducing a slit width, as there is no slit to close. Then an imaging system with an aperture at an intermediate focus would be called for – and would reduce the signal considerably.

EBIT and some Penning traps with their strong magnetic fields (and considerable stray fields) interfere with the workings of traditional photomultiplier tubes (that involve free electrons travelling from dynode to dynode), if the detectors are too close (for example, when mounted on a normal-incidence spectrometer of the typical V-geometry [239]. The problem can be alleviated by using the folding mirror that is available in some spectrometers, that is normally provided to couple a calibration light source to the same output light path. Reversing the “normal” light path can thus put the detector away from the strong magnetic field region.) Solid-state detectors and CCDs do not have this problem and can be mounted close to the light source if needed.

One more problem concerns the wavelength range to be studied with ion traps. All detectors have some sort of intrinsic noise. In a traditional photomultiplier tube, there is no clear threshold between electronic noise and regular photon signal. Furthermore, sensitivity to red light requires cathode materials with a low work function and consequently high thermal noise (to be reduced, but not completely avoided by cooling). EBITs have hot electron guns that produce plenty of red stray light not far from the actual trap region. Thus present EBITs have problems with observations in the far red or near infrared. Edge filters reduce the background, but also any signal in this wavelength range. A channeltron, an open photomultiplier for the VUV, in contrast to a regular phototube, operates in saturation, and a clean discrimination of electronic noise and photon signal is possible. Channeltrons may have dark rates of as little as 1 count per minute, whereas a dark rate as low as 1 count per second is achieved with photomultipliers only in the near UV/VUV, using special high-work function cathode materials. The quantum efficiency then typically is near 10%. Bi- and Tri-Alkali photocathodes for the visible range are available with quan-

tum efficiencies that are higher by almost an order of magnitude, but only at the penalty of a higher dark rate. The CCD, in some versions spanning the wavelength range from near-infrared to the soft X-ray range (though not uniformly) is a very interesting newcomer. It permits single-photon counting or long exposure times (limited by the need to correct for cosmic ray or similar interference events) at very low background. The latter is an interesting technical problem, as some noise is intentionally added during read-out to ensure statistical reliability. One of the drawbacks of CCDs is the cycle time required for read-out, which is only partly taken care of by binning and frame transfer operation modes. So far, CCDs seem to be extremely useful for spectroscopy, but are not necessarily advantageous for straight atomic lifetime measurements in the ms range that are of interest in the present context.

With X-ray detectors (available in sizes that make large solid angle detection possible) each signal pulse may be analyzed for its energy, and many false counts can be rejected, resulting in very clean data [242]. The high signal-to-noise ratio of X-ray measurements makes it much easier to obtain precision lifetime data in the X-ray range than in the visible [240].

3.4. Typical traps

There is plenty of information on conventional ion traps and their working principles available in the literature [70,129,219,258,259]. For a detailed coverage of many aspects of trap physics and a seemingly complete literature survey up to 1993, the reader is referred to the excellent book by Ghosh [115].

3.4.1. Conventional ion traps. Electrostatic (Kingdon) traps exploit the working principle of a cylindrical capacitor: a thin (30 μm diameter) wire at a negative potential is surrounded by a cylinder at a more positive potential. The contraction resembles a can with an insulated wire along the central axis of symmetry. Top and bottom lids of the can deform the trapping field to yield some repulsion and thus some trapping along the axis of symmetry, too.

In Penning traps a strong magnetic field makes the ions gyrate around the field lines (radial confinement). An additional superimposed electric field (positively charged end caps for positive ions) prevents ions from escaping along the field lines. Superconducting coils are very efficient for the production of high field strengths, and consequently they are often being used for Penning traps as well. They have the additional advantage that a cold magnet bore helps to maintain a superb ultra-high vacuum by freezing out any contaminants.

In the radiofrequency (RF) (Paul-Straubel-Langmuir) trap, an oscillating (many-kHz to MHz) electromagnetic field between a ring electrode and two end cap electrodes exerts an average force on ions inside. The quadrupole field can be aided by a small DC field that helps to push the ions in along the axis of symmetry. The ideal electrode surfaces would be hyperboloids, but a cylinder and two flat lids, sometimes made from wire mesh for optical access, do as well, though maybe less perfectly. Other designs use a sequence of three cylindrical rings on a common axis. Additional electrodes are sometimes introduced to shape the trap fields and to suppress unwanted harmonics. As

the ions experience periodic forces, there are many possible resonances that would heat the ions and thus eject them from the trap. Similarly to a storage ring where ions travel through a periodic lattice of fields, certain conditions (combinations of RF field amplitude and frequency, DC field amplitude, ion mass and charge) are good for trapping while others have to be avoided (or can be exploited to selectively remove specific ion species, by pushing them out of the region of stability).

3.4.2. ECRIS plus electrostatic ion trap. The Kingdon trap with its thin central wire has a $1/r$ electrical field strength and a logarithmic potential. The trap voltages are of the order of a few kV and can be switched on and off within about a microsecond. This simplicity is appealing for both, experiment and modeling, and a number of studies present calculations of typical ion trajectories inside such a trap [125,129,219]. Ion injection is regularly done at right angles to the central wire, but at some sideways distance. As the ions have about the same initial velocity, but are at very different positions and directions with respect to the trap center, only about half of those ions of an ion beam that happen to be inside the trap volume when the trap voltages are switched on will be trapped. For a trap of 10 cm diameter, this will correspond to an ion beam section of about 5 cm. Ions in instable orbits will lead to an enhanced ion loss initially, and this may cause problems for the evaluation of atomic lifetimes in the few-ms range. Trapping more ions will require either a larger trap (in which the particles move in a larger space – not good for observation) or higher ion currents at injection, possibly by pulsing the ion source.

Kingdon traps have been built and used by many groups. In the present context the most notable of these are C.E. Johnson's group at Raleigh, NC, studying atomic lifetimes in ions in charge states up to $q = 3+$ [38–40] and D.A. Church's group at College Station, Texas [53]. The latter extended the range of charge states to be studied considerably, moving one of their traps to Reno, Nevada, and combining it with the local ECR ion source (ECRIS) [189–191]. The whole system has been built to UHV standards. Ion storage times of many seconds are (purportedly) routinely achieved (although the latest publication on multi-charged ions states a value of just below a second). Ion loss rates are determined by switching the trap voltages off and then counting the ions that reach a detector outside. Operations with ions of a few keV energy and with ions of charge states up to $q = 14+$ are claimed to be straightforward, as is a signal level on forbidden transitions in the visible spectrum that is good enough for lifetime measurements of 1% precision within a couple of hours. However, only a few results, on forbidden transitions in ions of Mn, have been reported with this precision [191], while most data reported from this work are in the uncertainty range of several and up to 15 percent. The higher precision would be most interesting.

A very recent new design of an electrostatic trap, by the Zajfman group [67,270], may catch more ions from an ion beam, as it is based on a tube of about 40 cm length and injection along the axis of this tube. If the trap is closed when the first ions, after reflection by an electric potential at the far end, come back to the entrance port, twice the tube length is the measure of the “chunk” that is cut from the ion beam. The ends of this tube are not simply made

up by caps at high voltage, but consist of a sequence of apertures on graded potentials. The device has been conceived as an analogon to an optical resonator cavity, with ions running back and forth across the field-free center part of the trap and then bouncing back – with a little focusing action – from the end cap fields. The trap has been tested with various singly charged ions (both positive [67,270] and negative [261]) of 4.2 keV. The present restriction to singly charged ions relates to the peculiar detection system employed: If ions in the trap lose their charge (due to charge-changing collisions, molecular break-up or, for negative ions, autodetachment), they are no longer trapped. Those neutral atoms that happen to fly towards the far end of the trap may now pass freely through the “end cap” apertures and strike a microchannelplate, a highly efficient detector for keV particles. For the $1s2s2p\ ^4P_{5/2}$ level in He^- , the demonstration experiment yielded raw lifetime data that are statistically better than 1%. However, a correction for the influence of black-body radiation was necessary and amounted to about 15% (with some uncertainty), so that the final result of (343 ± 10) μs bears a 3% error bar. The result agrees well with that from earlier work at the Aarhus ion storage ring [2]. Moreover, the same data yield a first, similarly precise result for the combination of the much shorter, but apparently rather similarly-lived levels with $J = 1/2$ and $3/2$ ($\tau = 8.9 \pm 0.2$ μs).

While the builders of this trap have reported work on singly charged ions only, one may speculate about future uses with multiply charged ions. The height of the potential barriers (end caps) is a function of ion charge state q , since an energy qeU is needed to overcome a potential difference U . Starting out from multiply charged ions of a given ion energy that are trapped, sequential losses of charge by collisions may eventually lead to a charge state that is low enough so that the ion can pass the barrier and be detected. If measured step-by-step (charge state by charge state), it may be possible to see the influence of a particular long-lived excited level against a majority of ground-state ions, and thus to derive an atomic lifetime value. The technique yields a signal that is proportional to the number of ions that undergo small-angle scattering. Large-angle scattering events also cause losses from the sample, but are not detected.

3.4.3. Heavy-ion storage ring. The ultimate long trap to capture a section of an ion beam would be a ring vessel to thread the beam in (Fig. 2). This, indeed, is what is being done at heavy-ion storage rings [196]. As with yarn on a spool, the ion beam in the ring needs to be slightly displaced after a turn in order to accept the next turn. This technique, called stacking, permits the accumulation of a stored ion beam over about 30 turns (at TSR Heidelberg) before the phase space of the ring is filled. Next the ion beam can be cooled by interaction with a cold electron beam of about the same velocity, which shrinks the beam in phase space and would permit to add more of such accumulation cycles. “Cold” and “Hot” in this context refer to the irregular motion of the particles in the moving rest frame, motion that is superimposed on the high mean velocity of the ions. Thanks to the effect of *kinematic compression* [148], such a fast particle beam appears to be “cool” in longitudinal direction, if nothing but a constant kinetic energy has been

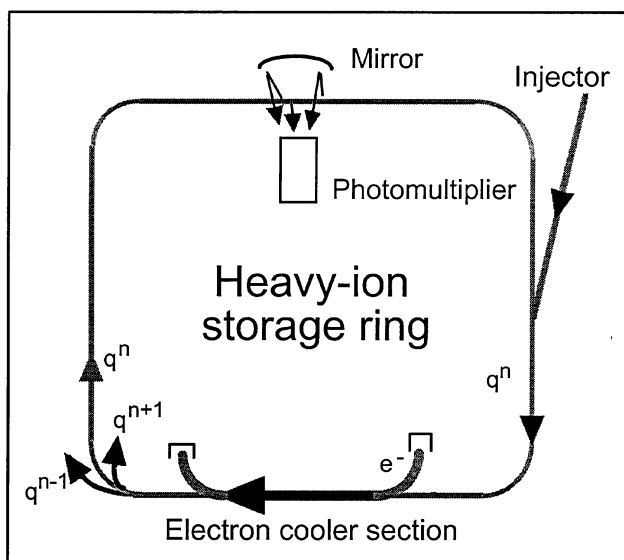


Fig. 2. Schematics of a heavy-ion storage ring with particle detection (bottom) and photon detection (top).

added by the acceleration process to the thermal motion from the ion/electron source. In contrast to the “cool” electron beam, the fast ion beam may have been stripped (changed in average charge state) by collisions with matter after acceleration of the originally singly charged (negative or positive) ions from the source, and it therefore is “hot”. Distant collisions of electrons and ions in the merged-beam section move some of the “heat” from the ions to the electron beam that is then deflected out and dumped. Because of intra-beam scattering, also the different thermal energies of longitudinal and lateral motions slowly equilibrate, and eventually also the lateral motion in the ion beam can be reduced by extended (repeated) longitudinal cooling.

Such a cooled ion beam is of utmost interest for many detailed atomic physics studies, like on dielectronic recombination or laser-assisted recombination, or as a reservoir to start state-selective laser excitation processes from. Unfortunately, hardly any of the original excited level population of the ions will be living long enough to be amenable to excitation by laser light after cooling, which may require a few seconds for low-charge state ions. The process is faster for highly charged ions, but there the energy intervals between atomic shells are far beyond the reach of lasers. Consequently the choice of options is limited to lifetime experiments that depend either on the initial excitation process (in the ion source or the injector) and no cooling, or use cooling and subsequent electron capture (in a gas jet target or in the electron cooler), unless the excitation interval is very small (as is the case for hyperfine structure in the ground state). Examples for these options are discussed below.

Storage rings come in many sizes. Some are second-hand (the CELSIUS ring at Uppsala had a first life at CERN), some are multi-purpose (ASTRID at Aarhus alternates as a storage ring for ions and as an electron ring for the production of synchrotron radiation), others were designed as test beds for bigger machines (TSR at Heidelberg for ESR at Darmstadt, but the bigger machine (circumference 108 m) was built before the smaller one (circumference 55 m) came to life), and some are the result of a small-is-beautiful idea: ELISA at Aarhus is a small offspring

of ASTRID, but dedicated to atomic and molecular ion storage. Oak Ridge once planned a full-size ion storage ring, but the project was canceled before serious construction started; Raleigh (North Carolina) was close to securing a storage ring project by financing it from sources that seemed safe from the swings of daily politics, but then in a federal election the local congressional seat fell to the other party and to a congressman of fervent disbelief in science. KVI Groningen tried for a small ring, based on their experience with ECR ion sources and RF quadrupole guide structures, but was denied funding. There are US users of the now solely European ensemble of heavy-ion storage rings who are lobbying for an American ring. The Chinese have a project that calls for a storage ring laboratory apparently modelled after GSI's rings (mostly intended for nuclear physics). However, for the time being, the rings of interest for atomic lifetime measurements are ASTRID, CRYRING, ESR and TSR, with some very different parameters.

Although there is a tandem accelerator at Aarhus that can provide multi-MeV ion beams, ASTRID is being served by an injector that runs up to 150 keV. Similarly, CRYRING has a low-energy injector, while in both rings the ions can be accelerated inside the ring. This saves costs for the injector, but it also limits certain experiments. TSR at the Heidelberg Max Planck Institute for Nuclear Physics was built as an addition to a nuclear physics laboratory that already had several conventional accelerators, of which a 2 MV single-stage and a 12 MV tandem accelerator, plus a linac that can act as a postaccelerator, nowadays serve the storage ring. This variety of injectors makes possible the injection of multicharged, multi-MeV ions that automatically are in an energy regime that permits better ion storage than at the multi-keV energies typical for traps that are fed by ECR ion sources directly. However, even a laboratory as complete as that at Heidelberg cannot fulfil all experimenter's dreams at once: While gas stripping in the tandem accelerator provides ample beams of ions up to charge state $q = 8$ or so [236], higher charge states need to be produced by a second stripping process after the accelerator. In case of Sc^{12+} , Ti^{13+} and Ti^{17+} [241,243] this double stripping reduced the fraction of ions available for eventual storage to about 6% of the ion source output beam. Of these ions, only a (unknown, but small) fraction will be excited. Compared to easily produced singly charged ions of some elements, the particle number stored therefore was down by more than two orders of magnitude. It then is no surprise that the results of these particular experiments were statistics-limited to a precision of a few percent, even after two full days of data collection. (In contrast, the high signal available with high ion currents sometimes leads to such small statistical uncertainties of the evaluation of individual data curves that one begins to worry whether the fraction-of-a-percent scatter of the results of a number of measurements is only statistical or bears the sign of some unknown foreign factor.) An RFQ structure (presently being tested as an additional injector) for the acceleration of multiply charged ions from a future ECR ion source promises to be a major improvement of this situation and will permit access to few-electron ions of the iron group elements that are of interest for understanding the solar corona, as well as to a few test cases of basic theory that are not well accessible any other way yet. Top of the line

(in terms of cost and maximum ion energy) is ESR at GSI Darmstadt, a ring that is fed from the heavy-ion synchrotron SIS and works at energies up to, say, 500 MeV/nucleon – perfectly suited for the long-term storage of almost bare very heavy ions.

There are interesting atomic physics experiments for each of these rings. The lifetime experiments of interest for this review cover intercombination, forbidden and hyperfine-induced transitions in a variety of ions of fundamental and astrophysical interest, with lifetimes in the 0.1 ms to 10 s range. I will not discuss the measurements on auto-detachment lifetimes of negative ions (these have been ably discussed elsewhere [2,3,5,6,121]), but only those on positive ions. Technical advantages compared to conventional ion trap measurements lie in the spatial separation of ion production and selection of a single isotope and charge state on one hand (in the injector and beam transport system), and storage (under UHV conditions at MeV energies) and detection on the other, which can be optimized separately. The result are reductions of the experimental errors often by more than an order of magnitude, and the outright removal of some sources of systematic error.

The uncertainties for atomic lifetimes in the millisecond range are largely statistics-dominated (the signal rate depends on the stored ion beam current), but for long (100 ms and more) lifetimes there also is the storage time correction problem. Total errors thus range from about 3% to 0.14% [78,235–237,241,243]. In these measurements, the stored ion beam current is monitored, for example, by a “beam profile monitor” that detects rest gas ions (with an electric field pulling those slow ions toward a microchannel plate (MCP)) that are produced by the interaction of the fast-ion beam with the residual gas. The rest gas ion signal normally demonstrates that the ion beam settles down sufficiently within about 0.3 ms or less. (This then is a practical lower lifetime limit of stored-ion beam experiments that depend on excitation taking place in the injector, and also the signal is to be followed for, say, at least 30 turns of the ions in the ring.) However, when varying the base pressure in the storage ring (by switching off some of the cryopumps, for example), the pressure may rise from 10^{-11} mbar to a few times this value, or even to 10^{-10} mbar. Under such circumstances and with the injection of strong ion beams (many μA current), the MCP-based detector at TSR has been found to yield extra variations of the ion current signal over times of about 50 ms which did not show in the optical signal and must be assumed to be artefacts of the rest-gas ion production and detection process.

Moreover, the storage ring cycle frequency (injection, observation, dumping) is typically a few Hertz for millisecond atomic lifetime studies. For example, the TSR ring at Heidelberg has a maximum cycle frequency of 5 Hz. It is not possible to extract a precise number for the beam storage time constant on-line from the 200 ms range of the beam current monitor (typical ion-loss time constants in the experiments mentioned above corresponded to ion storage lifetimes of 5 to 100 s). Instead, for the determination of this storage time constant, the beam current is being measured in between the optical data taking periods, with a slow-cycling mode of the storage ring, so that the signal of a pick-up antenna can be followed for, say, 30 to 50 s

and the effective storage time can be obtained with better precision. However, this system is not designed to cope with beam currents of several hundred μA as are available for some ion species, and not for the short-time injection process. While the frequency response of the electric probe system is appropriate for times of several seconds, the optical detection may not last longer than a fraction of the first second. Thus the measured beam storage behaviour from the slower, long-term device (on a time scale dominated by ground-state ions) may be different from the actual short-term behaviour (dominated by excited states of the ions and affected by the settling-down of the ion beam) that needs to be known in order to correct the optical data. This, in effect, is the major contribution to the assumed systematic error in some measurements of forbidden transitions [236]. Obviously, there is a need of further technical development. However, the total size of these corrections under the clean conditions of a storage ring usually is in the range of a few percent, and often as small as a tenth of a percent. Furthermore, there are indications that the apparent storage time depends on the tricks played to help the diagnostics systems pick up a signal (RF-bunching of the ion beam is done for the beam-life measurement, but this seems to reduce the storage time of the ion beam; without bunching the ion storage time approaches the theoretically predicted value, and so on), and that the actual ion storage under typical DC measuring conditions is more regular (as evidenced by the optical signal) than the electrical signal of the probes shows. Thus the corrections forced by the probe signal may be overestimates. Anyway, the above worries are serious only in the pursuit of even higher precision, and they concern the exact magnitude of corrections that regularly are much smaller (by order of magnitude) than the total errors of competing techniques.

The pressure correction in conventional ion traps is sought for by series of measurements at different pressures and an extrapolation of the results to zero pressure. With storage rings, each measurement is not only done at a much lower pressure, but also intrinsically referred to zero pressure by reference to the ion beam storage time constant. In addition, one can deliberately worsen the pressure (by switching off some pumps for a while) and check for the effect on ion beam storage and atomic lifetime result [241] and thus check the consistency of the correction process. However, the storage ring fails to operate at “high” pressures – pressures that are considerably lower than what is usually reached as the lower limit in conventional RF or electrostatic ion trap work.

Storage rings offer both optical and particle detection. Autodetachment of the “surplus” electron of negative ions, dielectronic recombination (DR) with electrons in the electron cooler, or ionization (effected by the cooler electrons or by collisions with the rest gas) have all been used for atomic lifetime measurements. The experiments rely on the fact that the storage ring parameters (magnetic fields) are optimized for a given charge state ion species. Ions that grabbed an electron and thus reduced their electric charge will be less deflected by the next bending magnet than the majority species (neutralized ions even fly on straight) and can be detected on the outside of the ring. Ions that suffered further ionization can be intercepted on the inside of the track (Fig. 2). The DR process has resonances so that

a state-specific detection process is possible and works very well for two-electron ions [215]. However, experience with four-electron ions [75–77] suggests that the variety of suitable atomic systems may be rather limited.

3.4.4. EBIT. Electron-beam ion traps (EBIT) combine the Penning trap principle with a strong, extremely well collimated electron beam along the magnetic field (of typically 3 T field strength B), thus also defining an axis of symmetry. The electron beam is compressed by the field, to a diameter of about 70 μm [247]. The “Penning” parts of the trap are completed by drift tubes on different potentials that keep ions in the trap volume axially confined. The electron beam serves several purposes: The electrons collisionally ionize atoms from the ambient gas (under UHV conditions) which then are confined by the trap. If collisions are sufficiently frequent (that is why the electron beam needs to be so tightly focused) and energetic, stepwise ionization to ever higher charge states can proceed, moderated by the interplay of ionization, recombination and charge-changing collisions with the rest gas (this indicates the need for UHV). The limit is given by the electron energy and the increasing ionization potentials of highly charged ions. The second job of the electron beam is a compensation of the space charge of the cloud of positive ions that is being built up in the trap. Even with a strong magnetic field for radial confinement, the ions would repel each other and move away from the location of the electron beam, if the attractive potential of the latter and the space charge compensation were absent. We will come back to these points and their importance for lifetime measurements below.

EBIT offers spectroscopical access to highly charged ions practically at rest, just by using a room-sized apparatus with some auxiliary equipment. The basic idea exploits the same collisional production of highly charged ions as the fast-ion beam work. However, one has to realize that in the interaction of a fast ion with a solid target, most of the collisions are with the electrons of the target material. Basic kinematics explains why then energies of 500 MeV/u [119] are necessary to reach bare uranium in a fast-beam experiment with a stationary target, while an electron beam of 250 keV, hitting a confined ion for some time, can reach the same charge state [183]. The difference in energies corresponds to the proton/electron mass ratio, while the collision velocity is the same. Of course, there are experiments that make good use of the fast-moving ions, but there are others that rather would want to avoid the problems of secondary X-ray and electron radiation, Doppler shift and Doppler broadening, and other side effects associated with very energetic ion beams. In this review, we stick to the latter experiments.

Atomic lifetime measurements with EBIT have been done in two very different lifetime ranges, femtoseconds (by a line-broadening measurement on Ne-like Cs^{45+} [18]) and milliseconds. In X-ray spectroscopic studies of fusion plasmas the spectral resolution has been increased far enough so that the Doppler broadening, caused by the thermal motion of the ions, could be evaluated as a temperature diagnostic in the keV range. In the Cs experiment with EBIT, in contrast, the temperature of the stored ion sample was reduced by evaporative cooling until the remaining ions showed less Doppler broadening than the natural line width

of a short-lived (femtosecond) resonance level. Evaluating the total line width, it was possible to extract the natural lifetime. This is a technological feat and a useful result. However, resonance transitions and allowed decays are close to the hydrogenic case that theory is covering sufficiently well, and in most cases there is little further interest (beyond the beauty of the demonstration) in the measurement of femtosecond lifetimes of highly charged ions, because all will be in the same ball park. It is difficult to envisage a precision lifetime measurement (to a few percent or better) based on X-ray line widths. We therefore return to the forbidden decay modes and their measurement.

EBIT has been designed for the production of highly charged ions, and in this it excels (see for comparison the above discussion of the multi-step procedure to produce a beam of highly charged ions for injection into a heavy-ion storage ring). However, many “real-world problems” in solar or plasma physics concern ions that by yesteryear’s standards were highly charged, but are not by present day EBIT capabilities. For example, the ionization potentials of many $n = 3$ spectra of iron group elements (and some way beyond) are in the 1 keV range, while typical EBITs are designed to work at electron energies of, say, 30 to (SuperEBIT) 250 keV. The electron optics design is far from optimum when run at energies of just a few keV, and much lower electron currents are realized in this range, much lower than under optimum conditions. Lower electron beam currents mean lower collision rates and consequently a lower signal, too. However, running at an electron beam energy that exceeds the ionization potential of the wanted charge state (in an attempt to force a higher signal rate by means of a higher electron beam current), removes (by further ionization) the ion species of interest from the sample, and the more-highly charged ions might even perturb the measurement of transient phenomena (like decay curves) by recombination [242] (Fig. 3).

Aiming at “low” charge states with regular EBITs therefore requires tricks, like an interruption of the stepwise ionization of Ar atoms at a time shortly after the start, so that the charge state distribution has not yet reached the final, steady-state equilibrium. The same incomplete ionization can be arranged for by worsening the ultra-high vacuum: More atoms are available for ionization and excitation by the electron beam, but then the conditions of the experiment are less clean than wanted. The cleanest way yet has been employed at the LLNL EBIT laboratory, where the electron beam formation has been done at voltages of a few keV, but then the drift tube potential has been set in a way to slow down the electron beam in the trap region, to energies as low as 90 eV. While this is still far from optimum running conditions, this technique offers the flexibility to check for the production threshold of unidentified lines in the spectrum, down to charge states of order $q = 3$ or $q = 4$. This is very helpful for spectra that are dominated by a number of unidentified lines [239,246].

Atomic lifetime measurements in EBIT rely on the sharp switching off of the electron beam [17,223], although early modulating techniques with a variation of the electron beam energy around the production threshold of a given charge state also turned out rather successful [229,256]. The physical difference is that there remains a partial space-charge compensation by the lower-energy, full-current electron

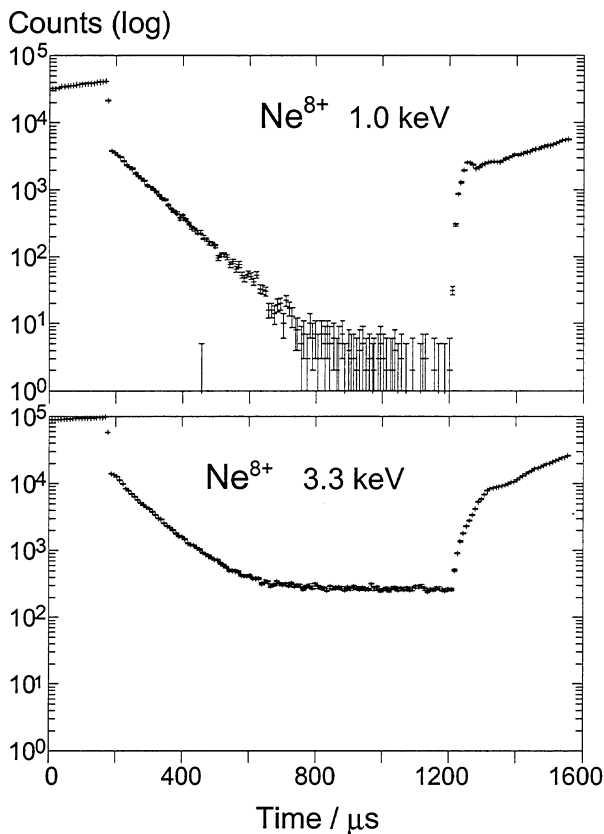


Fig. 3. Example of X-ray data curve. Decay $1s^2\ ^1S_0 - 1s2s\ ^3S_1$ in Ne^{8+} [242]. The signal rate at the lower electron beam energy – just sufficient for the production of the desired charge state ions – is lower than when working at a higher electron beam energy and current, but the decay curve is also much cleaner.

beam in the older technique, while in the new technique all further electron-ion collisions are intentionally avoided by lack of free electrons, at the cost of a change of geometry of the trapped ion cloud that expands radially to a new equilibrium. Consequently, there is no “rapid cycling” any longer, as the ion cloud does not overlap as well as before with the electron beam when that is switched on again. This is clearly evident in the decay curve data that show a slow recovery [224].

For the transition from steady-state conditions (with the electron beam “on”) to the magnetic trapping mode (with the electron beam “off”) one would ideally have a perfect switch that stops any further excitation and ionization and also removes the remaining free electrons from the trap volume in order to prevent further recombination. In practice, the electron beam energy and the electron beam current are best both reduced to zero within about 0.1 ms. Excitation, of course, stops when the electron beam energy drops below the excitation threshold, so lifetimes can in principle be measured to a fraction of that 0.1 ms total switching-off time. As most lifetimes measured on forbidden transitions in the visible are in the range of a few ms, such a switching time is, in fact, sufficiently fast for these. However, when the electron beam is being switched off, the trap electrodes may react to the sudden field and load changes by “ringing”. The dynamics of the change of geometry of the ion cloud and the switching transients overlap in time, and it has not yet been possible to study this transition period in detail. As it is now, one has to truncate the early data points until a curve with a stable exponential

behaviour is found. This problem presently limits the reliable and precise lifetime measurement using EBIT (in this way) to lifetimes longer than, say, 10 μs . It is conceivable that with some effort this range could be expanded to the sub-microsecond range, but for this atomic lifetime range other techniques, like recoil ion beams [134,135] (that also need further development work), may be easier to apply with confidence. However, it is exactly this range that is of interest for precision experiments on He-like ions (see below), and this makes any technique that promises reliable and precise lifetime data in this range worth pursuing.

At the other, long lifetime, end of the working range, EBIT also deserves development work. It has been established by Fourier Transform Ion Cyclotron Resonance (FTICR) frequency measurements [19] that in magnetic trapping mode EBIT stores ions for many seconds. If, however, atomic lifetimes in the second range are to be measured, the ion storage time constant has to be established with precision, as it will be a dominant correction for the optical signal. An interesting option to do this has been discussed in Peter Beiersdorfer’s group at LLNL: The lifetime predictions for the $1s2s\ ^3S_1$ level in He-like ions of O and Ne have been corroborated by measurements to better than 0.5% [57,242], and reliable calculations good to this level have been identified. Consequently, one might consider the 2 s lifetime of the same level in the Be^{2+} ion as known. An EBIT measurement of this lifetime could thus be turned around so as to derive the storage time constant instead of the atomic lifetime, possibly also as a check on the FTICR measurement. In principle, such Be ions could also be admixed to an ion cloud of primary interest as an on-line diagnostic tool for ion storage parameters. However, splendid ideas are not always perfect in practice: EBIT stores highly charged heavy ions better than light ions (at the same kinetic energy – from collisional exchange – the axial confinement barriers are higher for multiply charged ions than for ions in low charge states). This effect is employed in everyday EBIT operations, when a light-element gas (usually N_2) is bled into the trap to serve as a coolant by being evaporated from the stored ion cloud. An admixture of Be ions thus would yield only a lower limit to the storage times that are achieved with heavier element ions.

4. Atomic systems of interest

The present report discusses certain extensions of the older work as well as developments that came later than Church’s reviews [53,54] and that have led to some of the most precise atomic lifetime results ever obtained. The overall guideline of most of these experiments in their development phase has been to pursue “what can be measured” with a given ion production and light detection equipment, not always with an eye on cases that might be of interest for checks on theory. However, with increasing insight and improving equipment, one can also formulate clear strategies of covering atomic systems that are amenable to detailed theoretical study so as to render possible a meaningful match of experimental and theoretical studies.

The discussion below is structured into *intercombination transitions* (spin-changing electric dipole (E1) transitions) on one hand and (electric-dipole) *forbidden* magnetic dipole (M1) and electric quadrupole (E2) transitions on the other. There are good expositions of the particular problems of

theory with these transitions in the literature. For intercombination transitions in four-electron ions, see, for example, [143]; for forbidden transitions, see [11,12]. Within each of these categories, experimental progress is from basic systems to more complex ones (more electrons involved, or measurements on transitions between excited states), at the same time pushing the limits of experiment in order to intrigue similar pushes in theory. Recent experimental trends are discussed along the results, and a road map for further work will become apparent. Numbers to substantiate the discussions are given in the tables, for which I do not claim completeness. For example, wavelengths are not given for all transitions listed, because sometimes there are indirect observations, or only crude indications in the literature, or only calculated data available. For observations with filters such crude wavelength data are sufficient, while for work with laser excitation rather more decimal places are needed, but rarely to be found except by the very experiment. Remember, the long lived levels of present interest, because of collisional quenching, imply very weak lines in most light sources, so that in many cases they have not been seen in time for the classical spectroscopic data compilations. In a number of cases (for example, the N sequence in Table II and hyperfine-induced decays in Tables IV and V) I have chosen to present mostly or even exclusively calculated lifetime data – these cases will be tackled by experiment in the next few years, I trust. The reader will see from the wavelength ranges and lifetimes sought that some development work is necessary first.

4.1. Intercombination transitions

One- and two-electron ions have been studied elsewhere by a variety of techniques, aiming at the lifetimes of the $2s\ ^2S_{1/2}$ level in He^+ [207], the $1s2s\ ^1S_0$ level (two-photon decay) in Li^+ [206], or the $1s2s\ ^3S_1$ decay (M1 decay) discussed below, in the section on forbidden transitions. For higher- Z ions, less precise measurements used a variety of (mostly foil-excited) ion beams. Overviews of such beam-foil experiments on intercombination transitions can be found elsewhere [94,231,233,234,238]. However, hardly any of these qualify as precise lifetime measurements, data better than 5% being rare.

Beginning our systematic discussion with the fewest-electron systems, beam-foil spectroscopy has been employed to measure intercombination transition rates in He-like ions (mostly $n = 2$ levels) and for the intercombination transition decays of the $n = 3$ levels in Be-like ions (see [94] and refs. therein). The lifetimes involved, however, are much faster than appropriate for ion trap work, and rarely in the range of precision better than 10%. In contrast, Table I gives examples of recent ion-trap work on such intercombination transitions, and for several cases the precision reached is excellent.

The simplest systems that feature millisecond lifetimes and decays in the uv part of the spectrum, are the four-electron ions (Be iso-electronic sequence), with the $2s^2\ ^1S_0 - 2s2p\ ^3P_1^o$ intercombination transition. Early calculations of the rate of this transition scattered markedly,

Table I. Ions with intercombination transitions ($E1, \Delta S = 1$) of interest for ion trap lifetime measurements.

Ion	Level	Lifetime τ	Comment/Ref.	Wavelength λ (nm)
Be Sequence				
B^+	$2s2p\ ^3P_1^o$	(97.65 ± 0.5) ms	HSR [241]	267.79 nm
C^{2+}	$2s2p\ ^3P_1^o$	(8.3 ± 0.5) ms (9.714 ± 0.013) ms	RFT [164] HSR [78]	190.9 nm
N^{3+}	$2s2p\ ^3P_1^o$	(1.6 ± 0.2) ms	HSR [75–77]	148.65 nm
O^{4+}	$2s2p\ ^3P_1^o$	(0.5 ± 0.1) ms	HSR [75–77]	121.8 nm
B Sequence				
C^+	$2s2p^2\ ^4P_3^o$	$(6.8^{+0.43}_{-0.37}/86.2^{+12.6}_{-5.9}/19.5^{+1.3}_{-1.0})$ ms $(7.95 \pm 0.07/104.1 \pm 0.5/22.05 \pm 0.07)$ ms	RFT [95] HSR [237]	232.3–232.8 nm
N^{2+}	$2s2p^2\ ^4P_3^o$	$(1.0 \pm 0.06/13.4 \pm 0.97/3.2 \pm 0.23)$ ms $(1.32 \pm 0.05/13.9 \pm 0.2/3.3 \pm 0.07)$ ms	RFT [96] HSR [237]	174.6–175.4 nm
C Sequence				
N^+	$2s2p^3\ ^5S_2^o$	(4.2 ± 0.6) ms (5.7 ± 0.6) ms (5.4 ± 0.3) ms (5.88 ± 0.03) ms	RFT [159] RFT [146] EST [45] HSR [235]	214.0/214.3 nm
O^{2+}	$2s2p^3\ ^5S_2^o$	(1.22 ± 0.08) ms (2 σ) (1.25 ± 0.02) ms	RFT [144] HSR [244]	166.08/166.62 nm
Mg Sequence				
Al^+	$3s3p\ ^3P_1^o$	(0.300 ± 0.01) ms (0.305 ± 0.010) ms	RFT [145] HSR [241]	266.92 nm
Si^{2+}	$3s3p\ ^3P_1^o$	(59.9 ± 1.8) μs	RFT [162]	189.2 nm
Al Sequence				
Si^+	$3s3p^2\ ^4P_1$	104/811/406 μs	RFT [44]	234 nm
Si Sequence				
P^+	$3s3p^3\ ^5S_2^o$	(167 ± 12) μs	RFT [42]	221 nm
S^{2+}	$3s3p^3\ ^5S_2^o$	(48 ± 5) μs	RFT [124]	173 nm

EKT ECR ion source plus Kingdon ion trap, EST Electrostatic ion trap, HSR heavy-ion storage ring, RFT RF ion trap, PT Penning ion trap.

Table II. Ions with $M1/E2$ transitions of interest for lifetime measurements using ion traps. Iso-electronic sequences below Ar. Wavelength data are approximate from various sources.

Ion	Level	Lifetime τ	Comment/Ref.	Wavelength λ (nm)
He Sequence				
Li ⁺	1s2s ³ S ₁	49 s (58.6 ± 12.9) s	Theory [81] RFT [158]	21.0 nm
Be ²⁺	1s2s ³ S ₁	1.8 s	Theory [81]	10.46 nm
B ³⁺	1s2s ³ S ₁	(149.8 ± 0.45) ms	HSR [216]	DR
C ⁴⁺	1s2s ³ S ₁	(20.63 ± 0.05) ms (20.589 ± 0.042) ms	HSR [215] HSR [216]	DR DR
N ⁵⁺	1s2s ³ S ₁	(3.905 ± 0.05) ms (3.92 ± 0.13) ms	HSR [215] EBIT [17]	DR X-ray
O ⁶⁺	1s2s ³ S ₁	956 ± 5 μs	EBIT [57]	X-ray
Ne ⁸⁺	1s2s ³ S ₁	92.0 μs 91.58 μs (90.5 ± 1.5) μs (91.7 ± 0.4) μs	Theory [81] Theory [147] EBIT [256] EBIT [242]	X-ray
Mg ¹⁰⁺	1s2s ³ S ₁	(13.61 ± 0.49) μs	EBIT [229]	X-ray
Be Sequence				
Ar ¹⁴⁺	2s2p ³ P ₂ ^o	15.2 ms 15.6 ms (13.4 ± 0.7) ms	Theory [245] EKT [214] Theory [190]	590.19 nm
Ti ¹⁸⁺	2s2p ³ P ₂ ^o	1.0 ms	Theory [245]	228.90 nm
B Sequence				
Ar ¹³⁺	2s ² 2p ² P _{3/2} ^o	9.4 ms (8.7 ± 0.5) ms (9.12 ± 0.18) ms (9.7 ± 0.1) ms	Theory [49] EBIT [224] EKT [190] EBIT [22]	441.24 nm
Ti ¹⁷⁺	2s ² 2p ² P _{3/2} ^o	(0.627 ± 0.010) ms	HSR [243]	177.80 nm
C Sequence				
O ²⁺	2s ² 2p ² ¹ S ₀	520-545 ms (530 ± 25) ms	Theory [105,112,251] HSR [244]	232.16/436.45 nm
F ³⁺	2s ² 2p ² ¹ S ₀	298–310 ms (304 ± 5) ms	Theory [105,112,251] HSR [244]	187.58/353.33 nm
Si ⁸⁺	2s ² 2p ² ¹ D ₂	38 ms (38.3 ± 0.3) ms	Theory [251,112] HSR [236]	198.49/215.0 nm
Ar ¹²⁺	2s ² 2p ² ³ P ₂	44 ms	Theory [105]	833.96 nm
Mn ¹⁹⁺	2s ² 2p ² ³ P ₂	1.5 ms	Theory [105]	255.88 nm
Fe ²⁰⁺	2s ² 2p ² ³ P ₂	1.1 ms	Theory [105]	229.83 nm
N Sequence				
Na ⁴⁺	2s ² 2p ³ ² D _{3/2} ^o	41.6 s	Theory [188]	206.79 nm
Mg ⁵⁺	2s ² 2p ³ ² D _{3/2} ^o	9.58 s	Theory [188]	180.59 nm
Si ⁷⁺	2s ² 2p ³ ² D _{3/2} ^o	684 ms	Theory [188]	144.58 nm
Si ⁷⁺	2s ² 2p ³ ² D _{3/2} ^o	23.7 s	Theory [188]	144.06 nm
Si ⁷⁺	2s ² 2p ³ ² P _{3/2} ^o	23.8 ms	Theory [188]	94.92/276.39 nm
Si ⁷⁺	2s ² 2p ³ ² P _{3/2} ^o	9.7 ms	Theory [188]	94.44/272.36/274.21 nm
S ⁹⁺	2s ² 2p ³ ² D _{3/2} ^o	69.8 ms	Theory [188]	121.8 nm
S ⁹⁺	2s ² 2p ³ ² D _{5/2} ^o	2.87 s	Theory [188]	120.1 nm
S ⁹⁺	2s ² 2p ³ ² P _{1/2} ^o	5.28 ms	Theory [188]	79.16/226.14 nm
S ⁹⁺	2s ² 2p ³ ² P _{3/2} ^o	2.18 ms	Theory [188]	78.06/217.37/222.82 nm
Fe ¹⁹⁺	2s ² 2p ³ ² D _{3/2} ^o	720 μs	Theory [188]	80.44 nm
O Sequence				
F ⁺	2s ² 2p ⁴ ¹ S ₀	456 ms 435 ms (423 ± 10) ms	Theory [104] Theory [112] HSR [47]	224/429 nm
Ne ²⁺	2s ² 2p ⁴ ¹ S ₀	218 ms (226 ± 16) ms	Theory [112] RFT [69]	181.5/334 nm
Si ⁶⁺	2s ² 2p ⁴ ¹ D ₂	62 ms (63.6 ± 0.7) ms	Theory [112] HSR [236]	208/228 nm
Ar ¹⁰⁺	2s ² 2p ⁴ ³ P ₁	(14.8 ± 1.1 – 0.48) ms	EKT [268]	693 nm M1
F Sequence				
Ar ⁹⁺	2s ² 2p ⁵ ² P _{1/2} ^o	9.6 ms (8.53 ± 0.24 – 0.17) ms (8.70 ± 0.37) ms (9.3 ± 0.1) ms	Theory [49] EKT [268] EKT [190] EBIT [22]	553.4 nm
Ca ¹¹⁺	2s ² 2p ⁵ ² P _{1/2} ^o	2.1 ms	Theory [49]	332.67 nm
Sc ¹²⁺	2s ² 2p ⁵ ² P _{1/2} ^o	1.0 ms (1.00 ± 0.03) ms	Theory [49] HSR [241]	263.72 nm
Ti ¹³⁺	2s ² 2p ⁵ ² P _{1/2} ^o	0.54 ms (0.513 ± 0.010) ms	Theory [49] HSR [243]	211.73 nm
Al Sequence				
Mn ¹²⁺	3s ² 3p ² P _{3/2} ^o	(31.32 ± 1.82) ms	EKT [191]	

Table II. *continued.*

Ion	Level	Lifetime τ	Comment/Ref.	Wavelength λ (nm)
Fe ¹³⁺	3s ² 3p ² ² P _{3/2} ^o	(17.52 ± 0.29) ms	EKT [192]	
Si Sequence				
Mn ¹¹⁺	3s ² 3p ² ¹ S ₀	(1.5 ± 0.2) ms	EKT [191]	
Mn ¹¹⁺	3s ² 3p ² ¹ D ₂	(11.16 ± 0.10) ms	EKT [191]	
Kr ²²⁺	3s ² 3p ² ³ P ₂	(6.8 ± 0.1) ms	EBIT [239]	
P Sequence				
Ar ³⁺	3s ² 3p ³ ² P _{3/2} ^o	(243 ± 73 – 79) ms	EKT [268]	
Mn ¹⁰⁺	3s ² 3p ³ ² P _{3/2} ^o	(3.0 ± 0.2) ms	EKT [191]	
Mn ¹⁰⁺	3s ² 3p ³ ² P _{1/2} ^o	(6.17 ± 0.29) ms	EKT [191]	
Mn ¹⁰⁺	3s ² 3p ³ ² D _{3/2} ^o	(35.1 ± 1.43) ms	EKT [191]	
S Sequence				
Ar ²⁺	3s ² 3p ⁴ ¹ S ₀	(133 ± 24) ms	EST [209]	311 nm
		(159.7 ± 9.7 – 38.4) ms	EST [267]	
Mn ⁹⁺	3s ² 3p ⁴ ¹ S ₀	(2.1 ± 0.3) ms	EKT [191]	157/252 nm
Mn ⁹⁺	3s ² 3p ⁴ ¹ D ₂	(18.02 ± 0.16) ms	EKT [191]	296/420 nm
Cl Sequence				
Fe ⁹⁺	3s ² 3p ⁵ ² P _{1/2} ^o	(13.64 ± 0.25) ms	EKT [192]	

EBIT Electron-beam ion trap, EKT ECR ion source plus Kingdon ion trap, EST Electrostatic ion trap, HSR heavy-ion storage ring, RFT RF ion trap, PT Penning ion trap, DR Dielectronic recombination.

and the uncertainties of experimental (mostly beam-foil) lifetime data on highly-charged members of the Be iso-electronic sequence were at the 10% level and worse. The storage ring data on C²⁺ [78] and B⁺ [241] with their sub-1% uncertainties agree with the latest theoretical results from massively improved calculations (see [143]), but are more precise than any of them.

This seems a dry statement, but there is a lot more to it than appears at first sight. In fact, the same calculations, applied to the *resonance* transition (without spin change) of the same ions are claimed to be good to better than a part in thousand, and there is no experiment that comes anywhere close to testing this precision on lifetimes in the nanosecond and sub-nanosecond ranges. For intercombination transitions, the theory problem is much harder. In a non-relativistic calculation (multi-configuration Hartree–Fock (MCHF) with various relativistic extensions and corrections), which may make use of configuration interaction wavefunctions with thousands of components, it requires the precise calculation of a small term, the multiplet-mixing (spin-orbit coupling) matrix element, or the Breit term (there are various representations of different sophistication that are explained in the theoretical literature). In relativistic calculations (using multi-configuration Dirac–Fock (MCDF) algorithms), there are so-called large and small components to each wave function, and the intercombination transition probability turns out to result from the small leftover of the cancellation of two large, almost identical, components – again something very difficult to calculate precisely. The presently best calculations on Be-like ions claim an uncertainty of the intercombination transition probability of the order of 1%. This then is the (present) measure of quality and interest: Lifetime measurements on intercombination transitions that are more precise than 1% test theory.

There are just two data on four-electron ions that reach this mark, the aforementioned results on C²⁺ at 0.13% and on B⁺ at 0.5%. These experimental data corroborate several recent calculations (Fig. 4). Moreover, it appears that the theoretical error estimates have begun to shrink since the experiment confirmed that the mean values came

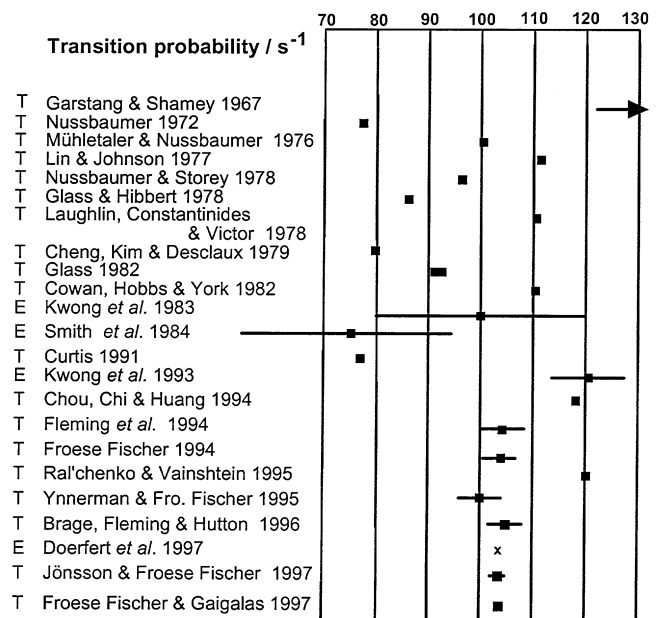


Fig. 4. Theoretical (T) and experimental (E) transition rate data (with year of publication) on the 2s² ¹S₀ – 2s2p ³P₁^o transition in C²⁺ (Be sequence). The experimental uncertainty of the heavy-ion storage ring result by Doerfert *et al.* [78] is as small as the center of the cross.

out right. However, it should be kept in mind that most of the precise calculations are not truly *ab initio*, but depend on slight adjustments (*fine tuning*) of calculated term intervals to experimental data. In fact, there were two slightly different theoretical predictions for the transition rate in B⁺ that referred to different experimental fine structure intervals. Fully independently of each other, improved wavelength measurements [173] and the lifetime measurements at the heavy-ion storage ring [241] settled the case and resolved the discrepancy.

Unfortunately, there are no precise lifetime measurements available yet for Be-like ions heavier than C. There are data for N³⁺ and O⁴⁺ that have been obtained at the heavy-ion storage ring, but with a non-optical technique. The experiment [75–77] had aimed at using the selective process of dielectronic recombination as a means to monitor the surviving fraction of stored ions in the 2s2p ³P₁^o excited level.

However, some technical parameters of the particle detectors available at the storage ring turned out to be counter-productive, and only an ionization signal was available in the end that yielded useable results, albeit not of notable precision. A vacuum ultraviolet detection system for TSR is presently in the planning stage that should permit to obtain precise lifetime data (near 1 ms) on these two ions in the not too distant future. Heavier ions will feature shorter lifetimes, lifetimes that are too short for storage ring experiments. Except for an old data point for Ne^{6+} that is based on an estimate (50% uncertainty) of data from a plasma experiment, all the other data, reaching from Fe^{22+} [136] to Xe^{50+} [193], are from beam-foil work on fast ion beams and carry uncertainties of order 10%. However, the same calculation that describes well the low- Z ions of the sequence [143], seamlessly matches a fully relativistic calculation at high nuclear charge Z [49]. It is also compatible with all the beam-foil data in that medium- Z region, and one may safely assume that it is more precise and reliable than all those experimental data beyond $Z = 6$ (C). As such good experimental data are available at low Z , and comparable data in the adjacent range are foreseen (see above), a “last” test of the calculations for transitions in the $n = 2$ shell of Be-like ions may be doable in the range near $Z = 10$ (Ne), which would suitably be done by a slow-ion beam technique [232].

This, however, is not to say that theory or experiment have reached the end of the line: It is quite conceivable to further improve even on the present excellent data on C^{2+} . In order to gain another order of magnitude in statistical significance, a factor of 100 in accumulated signal would be needed. This could, for example, come from a combination of multiple detectors (9 or 12 photomultipliers in a compact “star” arrangement seem feasible) and an extended collection time (3 weeks instead of 2 days). Of course, there are a number of technical and non-technical problems to be overcome. Besides the funding of the expensive low-noise detectors, such problems encompass the facing of ridicule and inertia of mind, as already encountered in comments like “there is no theory at that level – why bother?”. Correspondingly, I know of at least one theoretician who outlined his plan to improve theoretical precision by a factor of ten and was denied funding, because “there is no experiment to match”. Last, but not least, such precision is unknown terrain in ion beam diagnostics and storage ring behaviour, and there might be systematic errors that have not been recognized so far, but may well crop up and become important at the next order of magnitude in experimental precision. The presently available ion trap lifetime data on intercombination transitions are summarized in Table I.

4.1.1. Be-like to Mg-like ions. Be-like ions may be seen as two-electron ions, with the two electrons moving outside an inert core (consisting of the nucleus and the other two electrons). A comparison with data for other such effective two-electron systems might shed light on the validity of this picture, for example, after the Be-like ions a study of Mg-like ions. The Mg iso-electronic sequence features the same valence shell level structure as the Be sequence as far as levels are occupied, and it ought to cause comparable problems to calculate the $ns^2 \ ^1S_0 - nsp \ ^3P_1^o$ intercombination transition rate of electrons outside a core of closed shells, for $n = 3$

as for $n = 2$. However, in all such progressions one finds that the heavier ions have shorter-lived intercombination levels than the lighter ones (for numerical examples of two-electron systems, see [68]). This is likely caused by the penetrating s and p electron orbits that take little notice of inner electrons that may have larger angular momenta. Hence the electrons experience not the same core charge as indicated by the ion charge (plus one), but, with increasing atomic number, a less and less well screened nuclear charge. This somehow is also reflected in the level structure, where with increasing principal quantum number n of the valence shell the transition energy does not vary that much, and the transition wavelengths of similar transitions in analogous ions remain in the same wavelength range. In fact, the wavelengths of the $2s^2 \ ^1S_0 - 2s2p \ ^3P_1^o$ transition in B^+ and of the $3s^2 \ ^1S_0 - 3s3p \ ^3P_1^o$ transition in Al^+ are almost identical, as was exploited in the storage ring experiment [241]. The measured lifetimes, 100 ms and 300 μs , however, differ considerably, and the steep scaling precludes further investigations by the storage ring technique of the same transition in heavier two-electron ions. Similarly, there are rather precise lifetime data on the doublet-quartet intercombination transitions in B-like C^+ and N^{2+} ions (lifetimes in the 1 to 80 ms range) [237] that supersede earlier RF trap data, and it would be good to obtain storage ring lifetime data on the closely related case of Si^+ . However, according to the RF trap experiment by Calamai *et al.* [43,44] and to theory, all three lifetime components are below 1 ms, and thus there is little chance for a precision measurement by the storage ring technique.

I am not aware of any dedicated theoretical treatment of this regularity, with the exception of a study by Das and Idrees [68]. These authors give numbers for group IIIa – element ions B^+ , Al^+ , Ga^+ , and so on. Mind you, experiment [241] shows their transition probabilities for B^+ and Al^+ to be too low by a factor of 3 (or π ?), but that is no obstacle for deriving useful systematics once this shortcoming is known.

The sub-ms level lifetime in Al^+ comes close to the limits of the storage ring experimental technique. A comparison of experimental [241] and theoretical results indicates that this case is much less settled than the Be sequence. The scatter of even recent theoretical data suggests that the core is not only not inert, a fact modern calculations attempt to treat in a variety of ways, by core polarization models or extensive configuration interaction, but that there may be sheer computer size problems of calculations with as “few” as 10 electrons in closed shells.

4.1.2. More-than-two-electron ions. A different complication arises in the C iso-electronic sequence. The decay of the $2s2p^3 \ ^5S_2^o$ level to the $2s^22p^2 \ ^3P_{1,2}$ levels of the ground configuration of N^+ gives rise to two lines near $\lambda = 214$ nm. Here the $n = 2$ shell is the only open one, but there are more than two electrons that interact. The $2s2p^3 \ ^5S_2^o$ level is difficult to treat theoretically, as the usual optimization procedures (semi-empirical corrections) fail because of peculiarities of the wave functions. The storage ring results (5.88 ± 0.03 ms [235]) with their much higher precision (0.5%) lie outside of the 5% (1σ) error bars of both the latest other experiments and calculations. It is very good that theory has been developed to a stage where systematic

checks on the convergence and reliability of the calculations can be performed and are publicly demonstrated. However, it is amusing (for an experimentalist) to note that in the case of N^+ a calculation [128] was presented with the individual results of progressively more complex treatments, and that the second-but-last step turned out to be closer to the later experimental result from the heavy-ion storage ring than the last (that came close to an earlier result from an electrostatic ion trap [45]). Meanwhile several renewed theoretical efforts (not yet published) tend towards the most precise experimental number. Of course, this is a good sign, even as the new calculations come after the experimental fact. A very recent storage ring measurement on the same level in O^{2+} [244] corroborates the earlier RF ion trap results [144], but reaches a higher precision.

The heavy-ion storage ring measurements discussed so far covered test cases of a single intercombination decay, and all these systems are dominated by a single exponential decay component. A challenge to this technique is the three-component decay curve of the $2s^22p\ ^2P_{1/2,3/2}^o - 2s2p^2\ ^4P_{1/2,3/2,5/2}$ multiplet in the B-like ions C^+ , and N^{2+} , with predicted lifetimes (for C^+) that range from 5 to 100 ms. The experimental challenge is to obtain such statistically meaningful data that the extraction of three exponential components with reliable parameters becomes possible, from a common, spectroscopically blended, decay curve. By virtue of the matrix elements involved, the $J = 3/2$ level is always the most-long lived of such level multiplets, and its lifetime can be extracted from the slowly decaying tail of the data. At short and medium times after excitation, however, the $J = 5/2$ level component dominates because of the combination of high level population (approximately proportional to the statistical weight factor $g = 2J + 1$) and transition probability. The shortest-lived component, from the $J = 1/2$ level, begins with a lower intensity and dies out most quickly. This is aggravated by the time after excitation that elapses during ion beam transport, injection into the storage ring and settling down of the coasting ion beam. Even as this time – for intense beams of low-charge state ions – is only of order 0.3 ms [78,237], this is already a sizeable fraction of the lifetime of the $J = 1/2$ level in the case of N^{2+} . Comparable problems of ion cloud dynamics [157] may have put the earlier RF ion trap data in jeopardy: In the RF ion trap experiments by Fang *et al.* [95,96], the shortest-lived component in both C^+ and N^{2+} ions was obtained as very different from the theoretically expected value (in comparison to the other levels of the same multiplet) even as the signal statistics seemed quite good. A very recent storage ring experiment [237], however, reached even better statistics, and it was no problem then to analyze the data. In contrast to the earlier RF ion trap data, the newly measured lifetime pattern of the three levels in each ion matches some calculations that are not even very recent.

Interestingly, by the time the results of the heavy-ion storage ring experiment were being prepared for publication, experimenters at Caltech's Jet Propulsion Laboratory, using an electrostatic ion trap (Dave Church's solder set-up), claimed new results [227] to agree with the results by Fang *et al.* However, after being informed of the statistically extremely well-defined data from TSR, a new analysis was tried that assumed a longer settling period (2 ms instead of 1 ms). The analysis proved to be very sensitive to this

(a matter aggravated by insufficient statistics?), and the result for the $J = 1/2$ level lifetime component (which is near 8 ms) changed notably. It now is not any longer in gross discrepancy with the TSR data, while the other components remained unchanged at a distance of about two of their standard deviations. Such sensitivity of results to a parameter that is poorly controlled calls for caution.

Time and better experiments, on a number of systems, will tell in the long run which technique is the most reliable. So far, the heavy-ion storage ring is providing benchmarks for intercombination transition rates in low-charge ions, and conventional ion traps have a very hard time to reach and match that level.

4.2. Forbidden transitions

Forbidden transitions are those decays that cannot proceed by emission of electric dipole (E1) radiation, but only by the “normally” (in low- Z ions) much less probable magnetic dipole (M1) or electric quadrupole (E2) radiation. The forbidden lines most likely to be observable are the ones from levels in the ground configuration of an ion. These decays of the lowest excited levels can serve as a monitor of the presence of a given charge state ion in a plasma. If the transition rates are known, density measurements become possible (see [92]). For low to moderately high charge states, the fine structure intervals are small compared to the gross structure. Hence the wavelengths of such forbidden transitions in the ground complex even of highly charged ions may be in the visible or near-uv spectral ranges where interferometry is feasible and resolved line profiles [1] can shed further light on plasma processes. However, the fine structure intervals scale with Z^4 , that is steeply, along an iso-electronic sequence, and the wavelength change from one ion to the next may be drastic, so that not many members of a given iso-electronic sequence may be in the reach of a given detection system. Moreover, M1 transition rates scale with $(\Delta E)^3$, with ΔE being the transition energy, while E2 transition rates are lower at low Z , but then scale even more steeply, as $(\Delta E)^5$. Consequently there also is a “window” of elements in an iso-electronic sequence in which the atomic lifetimes can be measured by a given technique. Since the forbidden transition rates mainly depend on angular coupling factors (Racah algebra, atomic geometry) and the transition energy [62] (whereas allowed transitions depend on radial wavefunctions and on $(\Delta E)^2$), one can recognize as a rule of thumb that forbidden transitions with wavelengths in the visible or near-uv spectrum are typically associated with lifetimes in the range from milliseconds to seconds. L. J. Curtis has developed very involved systematizations [64] that combine fundamentals with spectroscopic energy information in order to yield, among other things, atomic transition rates. It will be interesting to see how these treatments fare in comparison to extensive computations when confronted with precise experimental forbidden-transition rate data.

For a great many ionic systems with not too many electrons and with nuclear charges up to $Z = 42$ (Mo), forbidden transitions in the ground and some excited configurations have been compiled by Kaufman and Sugar [149]. Their work relies on calculations that are compared to the much fewer experimental wavelength data. A most useful basis for a detailed systematization of those experimental wave-

lengths, which immediately relate to the level energies of the ground complex levels, had been laid by Edlén [85–90]. Useful in a different way are the survey calculations of level energies and transition rates (allowed and forbidden) in Li- to F-like ions given by Cheng *et al.* [49]. Astrophysical aspects have been collected, for example, in papers by Eidelsberg, Crifo-Magnant and Zeippen [93] and by Lynch and Kafatos [174].

Combining these sources, we have a good starting point for the exploration of forbidden transition probabilities in few-electron systems: The Cheng *et al.* transition rate data can be (slightly) corrected for the experimental term differences and then yield a reliable guide; if we then find agreement or less good agreement with sufficiently precise experimental data, we can contemplate where the shortcomings might come from. Of course, there are more theoretical data available, on individual iso-electronic sequences, from work that often was spurred by astrophysical interest. Of these calculations I want to name only a few and refer the reader to more references in the experimental papers cited. Be-like ions: [7,49]; B-like and F-like ions: [49,103,199,250]; C-like ions: [105,112,251]; N-like ions: [117,188,271]; O-like ions: [104,111,112]; Al-like ions: [132]; Si-like ions: [32,131,187]; P-like ions: [33,130,186]; S-like ions: [34]. This listing certainly is not complete, nor are the tables of experimental data (Tables II and III), but they will provide stepping stones for the interested reader.

A striking observation about the theoretical work is its complexity. As stated above, forbidden transition rates have been considered to be straightforward to calculate, as long as there was no experimental challenge to meet. For example, Curtis [62] demonstrated that in a single-configuration picture and at the non-relativistic limit the transition rate is a function only of geometry factors and transition energy and thus as precisely determinable as the transition energy.

Table IV. *Isotopes of Be sequence ions with hyperfine interaction-induced transitions. Examples are taken from the calculations by Marques et al. [181]. Wavelengths of $^1S_0 - ^3P_1^o$ transitions are experimental data, the wanted lines ($^1S_0 - ^3P_0^o$) are nearby at slightly higher wavelengths.*

Nucl. spin I	Z	λ/nm	Isotope	τ ($2s2p\ ^3P_0^o$)	τ ($2s2p\ ^3P_1^o$)
3/2	29	22.78	^{63}Cu	40.5 ms	9.26 ns
3/2	29	22.78	^{65}Cu	35.4 ms	9.26 ns
9/2	36	16.98	^{83}Kr	56.8 ms	2.44 ns
1/2	47	11.84	^{107}Ag	228 ms	741 ps
1/2	47	11.84	^{109}Ag	173 ms	741 ps
1/2	48	11.50	^{111}Cd	7.09 ms	687 ps
1/2	48	11.50	^{113}Cd	6.48 ms	687 ps
9/2	49	11.18	^{113}In	0.17 ms	639 ps
9/2	49	11.18	^{115}In	0.17 ms	639 ps
5/2	51	10.56	^{121}Sb	0.30 ms	558 ps
7/2	51	10.56	^{123}Sb	0.57 ms	558 ps
3/2	54	9.81	^{131}Xe	3.81 ms	464 ps
3/2	79	5.45	^{197}Au	2.59 ms	152 ps

Table III. *Ions with M1/E2 transitions of interest for lifetime measurements using ion traps. Heavy ions of low q/m , in iso-electronic sequences beyond Ar. Wavelengths are given only in those cases where direct observation is of interest.*

Ion	Level	Lifetime τ	Comment/Ref.	Wavelength λ (nm)
Ca ⁺	3p ⁶ 3d $^2D_{3/2}$	(1.113 ± 0.045) s (1.111 ± 0.046) s (1.17 ± 0.05) s	RFT [8] RFT [160] HSR [170]	
Ca ⁺	3p ⁶ 3d $^2D_{5/2}$	(1.054 ± 0.061) s (0.994 ± 0.038) s (0.969 ± 0.021) s (1.09 ± 0.05) s	RFT [8] RFT [160] RFT [211] HSR [170]	729 nm
Cu ⁺	3d ⁹ 4s 1D_2	(513 ± 60) ms	EST [209]	437.7 nm
Cu ⁺	3d ⁹ 4s 3D_2	(7 ± 14) s	EST [209]	380.7 nm
Kr ²⁺	4p ⁴ 1S_0	(14.8 ± 0.8) ms (13.1 ± 0.6) ms	EST [40] RFT [253]	350.5 nm
Kr ³⁺	4p ³ $^2P_{1/2}^o$	(47 ± 5) ms	EST [40]	322 nm
Sr ⁺	4d $^2D_{5/2}$	(345 ± 33) ms	RFT [113]	674 nm E2
Sr ⁺	4d $^2D_{3/2}$	(395 ± 38) ms (435 ± 4) ms	RFT [113] HSR [180]	687 nm E2
I ⁺	5p ⁴ 1S_0	(13 ± 1.3) ms	EST [46]	~ 340 nm
Xe ⁺	5d $^4F_{7/2}$	(57.2 ± 0.7) ms	HSR [169]	
Xe ⁺	5d $^2G_{9/2}$	(44.7 ± 1.1) ms	HSR [169]	
Xe ⁺	5d $^2G_{7/2}$	(3.4 ± 1.0) ms	HSR [169]	
Xe ²⁺	5p ⁴ 1S_0	(4.6 ± 0.3) ms (4.5 ± 0.3) ms	EST [40] RFT [253]	380.1 nm M1
Xe ³⁺	5p ³ $^2P_{1/2}^o$	(15.6 ± 0.9) ms	EST [40]	357 nm
Xe ³⁺	5p ³ $^2P_{3/2}^o$	(5.3 ± 0.5) ms	EST [40]	447 nm
Ba ⁺	5d $^2D_{5/2}$	(34.5 ± 3.5) s	RFT [175]	IR
Ba ⁺	5d $^2D_{3/2}$	(48.0 ± 5.9) s	RFT [156]	
Yb ⁺	5d $^2D_{3/2}$	(52.12 ± 1.00) ms	RFT [114]	435 nm E2
Yb ⁺	4f ¹³ 6s ² $^4F_{7/2}^o$	10.1 ⁺⁷ ₋₄ yr	RFT [212]	467 nm E3
Pb ⁺	6 $^2P_{3/2}$	(41.2 ± 0.7) ms	RFT [213]	
Hg ⁺	5d ⁹ 6s ² $^2D_{3/2}$	(9.2 ± 0.4) ms	RFT [140]	198 nm
Hg ⁺	5d ⁹ 6s ² $^3D_{5/2}$	(86.2 ± 3) ms	RFT [140]	1282 nm
Hg ²⁺	5d ⁹ 6s 3D_2	(34 ± 3) ms	EST [38]	217 nm

EST Electrostatic ion trap, HSR heavy-ion storage ring, RFT RF ion trap, PT Penning ion trap.

Table V. *Isotopes of Mg sequence ions with hyperfine interaction-induced transitions. Examples are selected from the calculations by Marques et al. [182]. Wavelengths of $^1S_0 - ^3P_1^o$ transitions are calculated data, the wanted lines ($^1S_0 - ^3P_0^o$) are nearby at slightly higher (not yet measured) wavelengths.*

Nucl. spin I	Z	λ/nm	Isotope	τ ($3s3p\ ^3P_0^o$)	$\tau(3s3p\ ^3P_1^o)$
3/2	29	36.47	^{63}Cu	39.0 ms	8.77 ns
3/2	29	36.47	^{65}Cu	34.1 ms	8.77 ns
9/2	36	25.73	^{83}Kr	55.6 ms	1.51 ns
1/2	47	17.46	^{107}Ag	211 ms	292 ps
1/2	47	17.46	^{109}Ag	159 ms	292 ps
1/2	48	16.97	^{111}Cd	6.50 ms	263 ps
1/2	48	16.97	^{113}Cd	5.94 ms	263 ps
9/2	49	16.46	^{113}In	0.157 ms	239 ps
9/2	49	16.46	^{115}In	0.157 ms	239 ps
5/2	51	15.56	^{121}Sb	0.27 ms	199 ps
7/2	51	15.56	^{123}Sb	0.51 ms	199 ps
3/2	54	14.32	^{131}Xe	3.43 ms	157 ps
3/2	79	7.98	^{197}Au	2.82 ms	44.7 ps

However, it is still a severe problem for theory to calculate fine structure intervals in complex ions precisely, often not getting closer to the experimental data than, say, 10%. (If experiment wants to find such a line, a prediction not better than to $\pm 10\%$ implies a similar wavelength interval – quite a search range for identifying a single line among typically hundreds! Tellingly, there is a paper [16] that presents improved fine structure intervals for exactly those four cases of a specific M1 transition in Ti-like ions for which experimental wavelength data were available at the time, but makes no prediction for others). Consequently, most calculations of forbidden decay rates nowadays refer to experimental energy data, correcting the calculations on the fly. This yields more reliable predictions, but it shows that theory is in dear need of improvement toward reliable precision. Also, most calculations nowadays are done with extremely large sets of wave functions in order to improve the calculated energy structure. This blurs the above “simple picture of forbidden transition rates”. However, if calculations refer to experimental transition energies and still yield answers that differ from each other and from experimental results, it becomes clear that good experiments test such calculations and provide benchmarks for future development – far beyond the aspect of collecting a few practical data for some or other terrestrial or astrophysical plasma application.

In fact, many astrophysical observations have such poor statistical significance that for their interpretation transition rate reference data good to, say, 10% are sufficient so as not to contribute markedly to the error budget. The physics goal of more precise lifetime measurements is set much farther: Precise calculations for everyday atoms in our environment, that is mostly neutral particles and a few singly charged ions, are very much more difficult than those for multiply-charged few-electron ions with a well-defined central potential. Also, extremely precise measurements as part of the chase for fundamental physics like parity violation require the study of heavy elements (because the non-classical effects are larger there), again a challenge for theory. Thus measurements on highly charged few-electron systems are one way to pursue quantum electrodynamical effects (for U^{91+} , theory

seems ahead of experiment in terms of precision), but most other work on multi-charged ions is providing stepping stones in the quest for precise work on *neutral* and *near-neutral* atoms, for which nowadays relativistic calculations are being done despite their need for extreme computer resources. Besides the fundamental physics quest for precise data on parity violation there is a more technical quest for better atomic clocks and frequency standards. These are expected from single, singly-charged, laser-cooled ions in traps. For the technical application the (very) long lifetime of certain levels is important, but it does not need to be known with any precision. However, imagine an atomic system in which from atomic clock development work the transition energy is known extremely well. It would certainly be very challenging to theory to meet such precision. An independent test would come from good lifetime measurements, as the transition rate implies details of the wavefunction that differ from those optimized for energy matching. In accordance with the different short-term goals, the following compilation is structured by discussing the forbidden transitions in few-electron ions first and then proceeding to the low-charge heavy ions.

4.2.1. *Few-electron ions.* 4.2.1.1. *He-like ions:* The simplest atomic system with a prominent M1 transition is the two-electron system with the transition $1s^2\ ^1S_0 - 1s2s\ ^3S_1$. The rate of this transition has been studied from neutral He (by laser absorption, the lifetime exceeding 1 h) to Xe^{52+} (by beam-foil spectroscopy, ps lifetime), that is for lifetimes that span 15 orders of magnitude (see [234]). In between these extremes, ion traps, a heavy-ion storage ring, an electron-beam ion trap, and a slow (recoil) ion beam have been employed, depending on the lifetime sought and the working range of the individual technique. The measurements at low Z were heroic, but did not produce very precise results. Similarly, fast-ion beams have problems near the long-lifetime end of their working range (a few hundred nanoseconds; measurements are available for $Z \geq 16$) and reach the short-lifetime end-of-the-range (a few picoseconds) at $Z = 54$, with a level of precision that does not surpass a few percent at best. The only precise data available are those in the range $Z = 5$ to $Z = 7$ obtained at the Heidelberg heavy-ion storage ring TSR [215,216] and those for $Z = 8, 10, 12$ obtained at the Livermore EBIT [57,229,242,256] (Figs. 3 and 5, Table II). The storage ring measurements employed dielectronic recombination, that is the electron cooler was set to merge the circulating ion beam with an electron beam of a specific, slightly different velocity. If the conditions were right on resonance, an electron might be captured while an electron of the ion might be excited, reaching a doubly excited atomic state. This could decay by the reverse process (not showing anything) or by radiative stabilization. Then the ion would have changed charge state and could be detected (Fig. 2). The pioneering experiment on C^{4+} reached an uncertainty of 0.2% – a true benchmark. Data for N^{5+} and B^{3+} did not quite reach this precision, and a recent attempt at measuring the multi-second lifetime in Li^+ is struggling to reach the 10% error range [263].

At EBIT, electron energy modulation near excitation threshold was used in their pioneer experiment on Ne^{8+} [256]. The signal was gathered by a large area X-ray detector

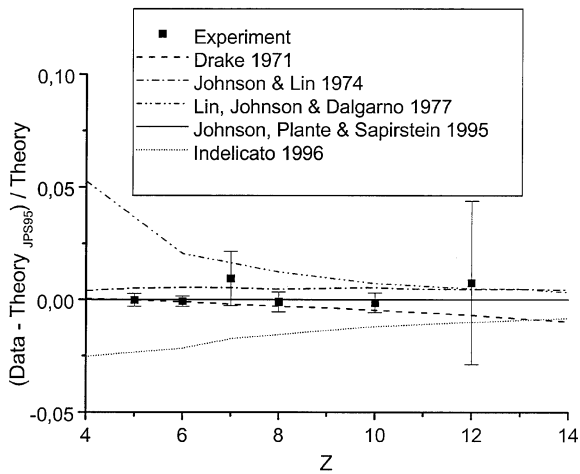


Fig. 5. Iso-electronic trend of the transition rate data for the $1s^2\ ^1S_0 - 1s2s\ ^3S_1$ magnetic dipole (M1) transition in He-like ions. Out of the full range of experimental data, $Z = 2$ to 54, only the range of the precision lifetime data is shown. Data for $Z = 5$ to 7 (leftmost data point) are from the heavy-ion storage ring TSR; the data for $Z = 7$ to 12 have been obtained at the Livermore electron-beam ion trap (EBIT). Additional (not yet published) EBIT data for N and F [197] confirm the trend of the data points with the small error bars. All experimental data and the results of theoretical calculations have been scaled by reference to the fully relativistic calculation by Johnson *et al.* [147]. (Full line, trace D).

that permits data acquisition in event mode and thus the on-line control of both, the light of the spectral line of interest as well as that of near-by spectral features. After developing the magnetic trapping mode [17] and finding agreement with the storage ring lifetime data on N^{5+} , new measurements have yielded lifetime data for O^{6+} [57] and Ne^{8+} [242] (Fig. 6) that are better than $\pm 0.5\%$.

A comparison with the various calculations shows (Fig. 7) that all but two calculations are ruled out by now. The only two calculations that agree with the precise data are those that employ “exact” wavefunctions, one starting from non-relativistic [81], the other from relativistic [147] wavefunctions. The non-relativistic picture is bound to fail at higher nuclear charges, and any measurement at the present level of precision, but at higher Z , is expected to prove this. However, the lifetimes of such ions (of order 1 μs and less) are too short for storage ring measurements, and also probably too short for reliable EBIT lifetime data. Slow ion beams seem most promising, and this brings us back to the aforementioned Ar^{16+} measurement on a recoil ion beam [134] and the improved detection system prepared by Schuch for an eventual measurement at Stockholm. Ten years after setting up, this still would be an up-to-date and worth-while experiment. For Ar^{16+} there are many calculations available, including at least one [172] that claims 0.5% precision. A future experiment ought to match this challenging mark.

4.2.1.2. *Be-like ions*: In a previous chapter I have discussed the measurement of the intercombination transition rate in Be-like ions, of the transition $2s^2\ ^1S_0 - 2s2p\ ^3P_0^o$, for example in B^+ and C^{2+} . While there are no forbidden transitions in the ground configuration of such ions (a singlet state gives rise to only one level), the excited triplet levels of the $2s2p$ configuration are very interesting (Fig. 1). The interest is in the $2s2p\ ^3P_2^o$ level that can decay either by

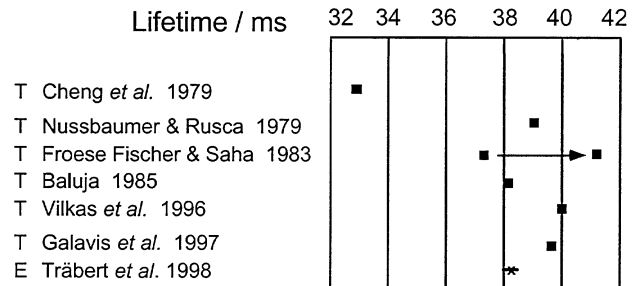


Fig. 6. Theoretical (T) and experimental (E) lifetime data on the $3s^2 3p^2\ ^1D_2$ level in Si^{8+} (C sequence). The arrow shows the change of an *ab initio* lifetime prediction when the calculated transition energy is *a posteriori* replaced by experimental data. The later calculations imply such a correction before publication.

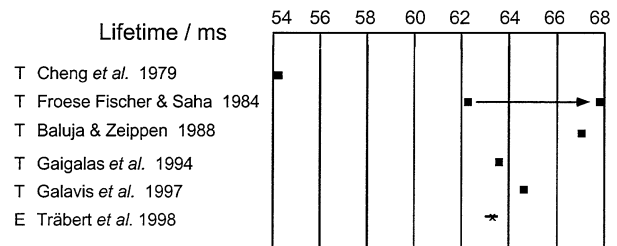


Fig. 7. Lifetime data on the $3s^2 3p^4\ ^1D_2$ level in Si^{6+} (O sequence). See Fig. 6 for comparison. Note that calculations by the same authors for Si^{8+} (C sequence) do fare differently in comparison with experiment.

magnetic quadrupole (M2) transition to the singlet ground state or by M1 transition to $2s2p\ ^3P_0^o$. (There also is a weak E2 branch to $2s2p\ ^3P_0^o$, but we neglect that one.) For low Z (C^{2+}), the M2 branch dominates and provides an interesting option for astrophysics [152], and it would indeed be interesting to check experimentally on the predicted 180 s lifetime – eventually, when that becomes feasible. However, for higher- Z ions of this iso-electronic sequence, the M1 decay dominates over the M2 branch, and a lifetime measurement of the $2s2p\ ^3P_2^o$ level would constitute a test of the prediction for the M1 decay rate in an excited configuration. Why care? The same predictions [49] that do reasonably well for forbidden transitions in the ground complex – as judged by the calculated versus measured energy intervals – do much less well for the excited state. This may simply be a shortcoming of the old calculation with too few configurations (Edlén cautions of intricacies of the wave functions [90]), but it may also be a more serious problem. A variety of calculations (among others, [7,199,214,245]) are available, and all of them need semi-empirical adjustment. The latest calculation, by Safonova *et al.*, comes close to experimental energies even at the *ab initio* stage. After the adjustment, the scatter of the predicted transition rates is of the order of a few percent – but the only measurement, on Ar^{14+} , by the combination of ECR ion source and Kingdon trap [189], deviates from theory by much more. It ought to be checked by independent measurements whether this deviation is the fault of experiment or theory. Future experiments might use EBIT (on Ar^{14+} , a precise wavelength measurement of the $2s2p\ ^3P_1^o - 2s2p\ ^3P_2^o$ transition has already been done using the Oxford EBIT [30]) or a storage ring (for example, on Ti^{18+} , after the implementation of the new injector at TSR).

The $^3P_0^o$ level is also interesting. In even isotopes, this level does not have a single-photon decay, and its predicted life-

time in C^{2+} is 3 years [166] – among other major problems, this lifetime is a bit too long for a graduate project. We will revisit this case below, in the context of hyperfine interaction.

4.2.1.3. *B- and F-like ions*: Both, B- and F-like, ions have a $^2P^o$ ground state (only the $J = 1/2, 3/2$ level sequence differs), giving rise to a single fine structure transition that is prominent in solar spectra and which thus provided an early incentive for Edlén's systematizations [87,61]. The same prominence makes these ions attractive for lifetime measurements. Such have been tried using the combination of ECR ion source and Kingdon trap [189,266–268], EBIT [224] and the ion storage ring [241,243]. The results (Table II) are intriguing. While the results of the former techniques agree with each other at the typically 5% uncertainties reached, and deviate from theory, the storage ring results (at 3% and 2% uncertainty, respectively), perfectly agree with theory for the B-like ion Ti^{17+} and come very close to theory in the case of F-like Sc^{12+} and Ti^{13+} . My personal conclusion from this is that the same calculations that are good for B-like ions are not quite as good for the additional electrons of the F-like ions (underlining the complexity of the purportedly simple problem of M1 rates), and that the mentioned non-storage ring experiments may be suffering from as yet unrecognized systematic shortcomings. The very latest (not yet published) data from a new, dedicated lifetime experimental set-up at the Livermore EBIT come close to a 1% uncertainty for both Ar ions [22] and also corroborate theory, in contrast to the earlier NIST EBIT and Kingdon trap results.

4.2.1.4. *C- and O-like ions*: C- and O-like ions are another likely pair of ionic systems with great similarities. The ground state electron configurations are $2s^22p^2$ and $2s^22p^4$, respectively, with a 3P ground term (J-values ordered as 0,1,2 and 2,1,0, respectively) and both a 1D_2 and a 1S_0 level in the same configuration. The latter has roughly twice the excitation energy of the former, and there are M1 and E2 decay branches. For example, the 1S_0 level can decay to 1D_2 (pure E2 decay), to 3P_1 (M1) and to 3P_2 (E2). Interestingly, for low charge state ions the E2 decay branch of the 1S_0 level to the while for higher charge states the M1 decay to the 3P_1 level outweighs the others by orders of magnitude. In an early calculation of this decay, by Froese Fischer and Saha [104], the uncertainty of the series expansion for the E2 decay in the O-like F^+ ion rendered its amplitude uncertain by a factor of two. A very recent storage ring measurement [47] fixes the lifetime (near 425 ms) to $\pm 2.5\%$ – quite a stringent test, and the result is only just in agreement with recent calculations by Galavís *et al.* [112]. Corresponding measurements on C-like O^{2+} and F^{3+} have very recently been done as well [244].

For the lower-lying 1D_2 level, the wavelength “window” of the experimental set-up at the TSR storage ring is reached only for more highly charged ions. The lifetime measurements yielded $\tau = 38.3 \pm 0.3$ ms for in the C-like ion Si^{8+} and $\tau = 63.6 \pm 0.7$ ms for the O-like ion Si^{6+} , that is both with a precision near 1% (Figs. 6 and 7). This is clearly less than the scatter of the more recent, quite extensive calculations [12,49,104,105,112,188,251] and underlines the need for further theoretical efforts.

4.2.1.5. *N-like ions*: A study of test cases in the N sequence would be interesting, too [117,188,271], in particular as N-like ions are a little bit more complex than the above ones, with a $^4S_{3/2}$ ground term in the $2s^22p^3$ ground configuration and $^2P^o_{1/2,3/2}$ and $^2D^o_{3/2,5/2}$ levels in the same ground complex. By virtue of the level structure, the $^2D^o$ levels are much more long-lived than the $^2P^o$ levels. In S^{9+} , for example, the lifetimes are expected in the few-ms ($^2P^o_{1/2,3/2}$), many-millisecond ($^2D^o_{3/2}$) and few-second ($^2D^o_{5/2}$) ranges, respectively. The level pairs lead to transitions with wavelengths that will not be spectrally resolved in experiments using filters. Consequently, the expected decay curves will have two components each that need to be fitted simultaneously. Table III so far has only theoretical lifetime entries, showing the range of values to be expected. Experiments with the existing set-up at TSR (UV range only) in a first round aim at the few-ms lifetimes of the $^2P^o_{1/2,3/2}$ level lifetimes in S^{9+} [244]. The interesting multi-second decays of the $^2D^o$ levels in the same ion, however, will have to wait for a VUV detector that can be coupled to the storage ring vessel without a material window.

4.2.1.6. *Al- to S-like ions*: In the ground complexes of these $n = 3$ shell ions, the same level structure as in B-like to O-like ions is repeated. There is the additional challenge for theory to deal with another closed shell, and there is the astrophysical aspect of the appearance of such ions of iron group elements in stellar plasmas. This has incited a series of measurements at Reno, in particular on Mn ($Z = 25$), using an ECR ion source and a Kingdon trap [189–191]. As the authors, Moehs and Church [191], state, the comparison of the experimental results (some claimed with errors as small as 0.9%, some with 15% uncertainty) with theory is inconclusive.

More dedicated measurements will be needed to clarify the situation. Perhaps it would be advisable to measure some ions by different techniques, in order to test and ascertain the reliability of the experimental techniques used. One such comparison has been tried by working with EBIT under different conditions. The subject of the studies was the $3s^23p^2$ 3P_1 level in the Si-like ion Kr^{22+} [224,239]. The measurement at the NIST EBIT reached an uncertainty of 9%, the one at the LLNL EBIT reached 5%. Both fell short of the predicted lifetime by more than their (1 σ) error bar. The NIST measurement used electron beam energies far above the value needed for the purpose (in order to increase the signal rate by means of the higher electron beam current) and also varied the gas pressure to exploit a transient ionization state for the measurement. The LLNL measurement worked just above threshold, but found that also for an electron energy just below the production threshold for the ion of interest there was some contaminating decay that spoils the hoped-for precision. Meanwhile, data recorded with much higher spectral resolution (using a transmission grating spectrometer [198] at LLNL [21]) indicate the presence of near-by unidentified lines. In fact, there are some 20 fairly prominent lines in a wavelength interval of about 10 nm near 384 nm – several of them probably from low charge state ions, and many of these identified tentatively (and by charge state only) at best, as well as some unidentified lines from highly charged ions, – and a continuum that appears like

an unresolved molecular band, but seems to be excited only at higher electron energies, possibly in synergy with the forbidden line of interest in the highly charged ion. If funding was available, this would be a truly fascinating spectroscopic and collisional physics problem worth studying. As it is, it is a nuisance that shows how purportedly simple experimental situations can turn out to be frighteningly complicated when scrutinized in detail. Truly clean conditions are difficult to achieve – and relatively best at the heavy-ion storage ring where only a single ionic charge state is being stored and under very good vacuum conditions – but then, sufficient ion beam currents of excited ions in these high charge states, combined with decent spectroscopic overview and resolution, are not available at any present storage ring.

By the way, EBIT is very good for identifying the charge state of unknown forbidden lines, as photon spectra at various electron energies clearly show when the excitation energy permits the production of a new charge state ion [60]. However, even in the UHV system of EBIT there are contaminations (with atomic species previously used), and there are ionic systems for which no theoretical data are available to proceed with line identification – for example in the iso-electronic sequences beyond Ar. Last, but not least, there are problems as basic as that of reference lines in the rare-gas spectra. There are prominent spectral lines of wavelengths known (since the 1930s) to six or seven decimal places – but their identification (beyond atomic species and, perhaps, charge state) remains unknown. Experienced spectroscopists at NIST say that they find such mystery lines in many elements and can't assign them. Atomic spectroscopy – a closed file?

4.2.1.7. *Ti-like ions*: In the discussion above I stated that fine structure intervals increase rapidly with ionic charge. The exception that “proves” this rule has, after a long search [97], been found in the Ti iso-electronic sequence. The $3d^4 \ ^5D \ Z=2-3$ fine structure interval is almost constant over a fairly wide range of ionic charges, rendering this fine structure transition a most interesting candidate line for plasma diagnostics purposes [1] by possibly using many different elements. The NIST EBIT has been employed to identify this line in four elements [194,221] (this work was then carried on at other EBITs and has reached up to Au^{57+} [239]), as well as determining the transition rate in Xe^{32+} . That result, a lifetime value of 2.15 ms with a 7% uncertainty [223], comes close to that of a (semi-empirically scaled) calculation. It also underlines EBITs capability for reaching high charge states and for measuring ms-lifetimes to some precision. That combination is presently unique.

4.3. *Heavy, low-charge ions and atomic clocks*

Low-charge state ions, preferentially of rare gases, are the most convenient ones to produce and excite in an ion trap. It is less easy to produce charge states higher than $q=2+$ by electron impact using a standard electron gun or laser ablation, although it has been done [40,163,209,255]. Next best choices are the molecular atmospheric gases (N_2 , O_2 , CO_2) or materials with a high vapour pressure (Hg). These boundary conditions describe much of the element choice in early lifetime work with conventional

(RF and electrostatic) ion traps that has already been compiled by Calamai [41] and reviewed by Church [53]. However, there are a number of physics-oriented studies that had other elemental priorities [8,23,25,26,29,36,98,126,185,217], and the quest for better frequency standards introduced elements chosen for their atomic structure rather than the ease of production.

Much of the ion trap work on singly charged heavy ions (Table III) was and is directed at measuring atomic masses, with notable success [72,73,142]. Other experimenters concentrated on spectroscopy [see [254)], and some on atomic lifetimes, though often only as a tool when determining collision rates, or as demonstration milestones on the way to better frequency standards and future atomic clocks. For example, if only a single ion is stored in a trap and the ground state resonance transition is being driven by laser light, there are ions that suddenly stop fluorescing, because the electron may no longer be available, having reached a metastable level (via another atomic decay branch) instead of the ground state. Excitation energy thus is being stored (“shelved”) until the ion undergoes spontaneous decay of that metastable level, and then the process can begin again. As the (resonance line) fluorescence is being switched on and off by the atom, the catch-phrase is *telegraphy mode*. From the statistical distribution of the lengths of the “off” time intervals one can construct a decay curve of the (not directly observed) metastable level – in some cases even of several levels, see the showcase demonstration of laser-cooled Hg^+ ions [140,141]. Fascinating as these studies are, the lifetime information on those “other” levels is not yet precise, but might well be made precise by dedicated future experiments.

In some cases the lifetimes of the metastable levels are so long that even hardy experimenters would get bored by waiting for the reappearance of the fluorescence in telegraphy mode. They then interrogate the ion by laser light of a different wavelength and shift population from the metastable level to some other level that does decay within a reasonably short time. In this way it was first checked that the Yb^+ level envisaged as extremely narrow (because it is so long-lived) does live for eight days at least [167], and meanwhile the same level has an experimental lifetime tag of order 10 years [212], though not a precise one. Let us return to lifetimes that are closer to the main body of work.

Among the ions studied most is Ca^+ , which has served as a test bed for trap physics, collision studies, high-resolution spectroscopy and the like. In the course of collision studies, the atomic lifetime was measured, by a number of groups [8,160,211], to some precision – but not always in agreement with each other. At long last, a different experimental technique has been applied that agrees best with some of the earlier data, but introduces new questions as well. The new technique is the heavy-ion storage ring (instead of Penning traps), in this case CRYRING at Stockholm [170]. With such heavy ions and low charge states, the energy of storable ions is far below the MeV range discussed above. This implies larger collision cross sections than at higher energies and a shifted balance of electron loss and recombination. In one case it was even found that an excited level in Xe^+ (at an ion energy of 40 keV) appeared to have a smaller destruction cross section than the ground state [179].

CRYRING has several ion sources, for singly or multiply charged ions. Taking advantage from their earlier experiments, experimenters there do not rely on observing the spontaneous decay of ions that might be still excited from the ion source, but actively influence the level populations by collinear laser light. Earlier on this technique has been demonstrated to resolve hyperfine structure [169,178].

Low charge state ions of heavy atoms are of interest to astrophysics. In many stellar spectra, just a few lines of a given singly charged species are available to obtain elemental abundances (and to charge heated discussions about stellar models and peculiar stars). There are many practical problems, however, with laboratory data on such level lifetimes, as the laboratory spectra often are very line-rich, requiring laser-spectroscopic techniques, while the transition rates of interest are in awkward ranges for laboratory work (though not necessarily in the range suited for ion trap work). Just to establish the context, I would like to mention as an example the work that is being done at Edmonton, with lasers on ion beams: Nanosecond lifetimes of Ca^+ have been measured to 1% precision, but the results differ from theory by 9% [118]. Among the many studies in the astrophysical context there is also work on Fe^+ [120] and on Yb^+ [205] – in the latter case there is laser excitation that starts out not from the ground state, but from the extremely long-lived level of atomic clock and ion trap interest.

While some ions have found so much interest that rather detailed studies have been performed, this matter in general is far from routine operation. For example, it is easy to obtain I^+ ions, which feature a transition that should show a lifetime of order 11 ms. However, nearby there is a line of I^{2+} that is also long-lived. Attempts using different filters for detection of the light from an electrostatic ion trap [46] and thus trying for a differential measurement did show a slight (3%) variation of the apparent lifetime when using the one or other filter which would enhance or reduce one of the contributions. However, the available filters are not narrow enough to suppress one of the lines completely. This puts the problem back to an evaluation of the relative production cross sections of singly and doubly charged ions of iodine, which were not known under the given conditions. A storage ring experiment would take the problem by the root, as only a single charge state ion would be stored and observed. However, because of the low charge-to-mass ratio, the ion energy accommodated by a ring like TSR would have to be lower than 1 MeV, and thus the storage conditions would be less clean than for the light ions for which precision lifetime results have been obtained.

4.4. Hyperfine interaction

So far I have presented spin change (intercombination) and higher-order multipole radiation (M1, E2) as mechanisms behind the “slow” decay of long-lived excited levels. Let us now turn to a third process, transitions that are mediated by a coupling of the electron shell with nuclear momenta, that is by hyperfine interaction. The classical case is the $1s2p\ ^3P_0^o$ level in He-like ions that normally cannot decay to the $1s^2\ ^1S_0$ ground state because absolutely no single-photon 0-0 transitions are allowed. However, with a non-vanishing nuclear spin, there may be hyperfine components that have the same total angular momentum quantum number

$F = J$ (electron shell) + I (nucleus). Then a hyperfine structure sublevel of the $^3P_0^o$ level mixes with a hyperfine structure sublevel of the $^3P_1^o$ level, which in turn mixes with the $^1P_1^o$ level (multiplet mixing). Obviously, the $^3P_0^o$ level is still longer lived than its partner level $^3P_1^o$, which in turn is longer lived than the $^1P_1^o$ resonance level. Beam-foil spectroscopy has demonstrated and used this effect in various He-like ions ([35,82,83,226], for further references, see [234]). Of course, one may claim the hyperfine structure effect to be known and turn the problem around. Then the lifetime measurement reveals the amount of mixing as a measure of the fine structure intervals that are not known from spectroscopy. Where available, spectroscopic data demonstrate that the exact position of the $J = 0$ level is still a problem for atomic structure theory [28].

In ions without a change of principal quantum number, that is in the Be and Mg iso-electronic sequences (and so on), the transition energies are smaller and thus also the resonance and intercombination transition rates. Hyperfine-induced transition rates will be correspondingly smaller yet – too small for beam-foil work, but probably just right for EBIT and the like. Marques and Parente [181,182] have calculated a number of examples of Be- and Mg-like ions (Tables IV and V). The details of their calculations might clearly be developed further than was done for their pilot study. However, their data show many interesting things: There are lifetimes in the ms to many μs range for Be- and Mg-like ions up to uranium – clearly a case for EBIT. Most of the transitions are in the EUV, requiring development work to combine sufficient spectral resolution, detection efficiency and time resolution. Amusingly, for the same isotope, Be-like and Mg-like ions have about the same lifetime – providing a handle for a systematic test of the experimental technique.

There is (at least) one heavy ion, In^+ , that features a hyperfine-induced decay in the working range of existing equipment (Table VI). This ion has been discussed repeatedly in the context of atomic clocks. It has a $5s5p\ ^3P_1^o$ level with an intercombination decay that is a bit too fast for ion trap work ($\tau = 440 \pm 40$ ns [202]), but it also has a $5s5p\ ^3P_0^o$ level with a hfs-induced decay at a wavelength near 236 nm [165] and a lifetime of (140 ± 20) ms [202,203]. A precise lifetime measurement calls for a storage ring in this case, even as those are not designed to handle singly-charged heavy ions at MeV energies, nor are multi-MeV rings equipped to handle sub-MeV ion beams. However, I trust that, for such an attractive case, the experimental questions (ion source, ion energy, available currents) will be sorted out in due course.

Let us now turn to the opposite problem, from a singly charged heavy ion to a heavy ion with only a single electron, $^{209}\text{Bi}^{82+}$. The proto-typical one-electron system, hydrogen,

Table VI. Hyperfine interaction-induced transitions in low- q/m ions.

Ion	Level	Lifetime τ	Comment/Ref.	Wavelength λ (nm)
Cd I sequence				
In^+	$5s5p\ ^3P_1^o$	(440 ± 40) ns	$E1/\Delta S = 1$ [202]	230.6 nm
In^+	$5s5p\ ^3P_0^o$	(140 ± 20) ms	hfs-induced [202]	236.5 nm

has a nuclear spin, and thus the ground state feature hyperfine splitting. The spin-flip between the two hyperfine levels gives rise to the 21-cm line of radio-astronomy and to the hydrogen maser radiation. Because of Z^3 scaling, the same transition in hydrogen-like $^{209}\text{Bi}^{82+}$ is in the near ultraviolet. This has been confirmed by a laser-resonance experiment on H-like Bi ions circling in the heavy-ion storage ring ESR at GSI Darmstadt [155]. The actual value of the transition energy, however, slightly differed from expectation and lead to the need for improving the nuclear model, for example assuming a “soft” distribution of the nuclear magnetic moment. Similar corrections were necessary for the interpretation of wavelength data that were obtained on other isotopes using EBIT [58,59]. The unique contribution of the storage ring, however, is the capability for lifetime measurements. After pulsed laser excitation, the fluorescence was followed and the decay constant determined. While in the first attempt a lifetime value was found that significantly differed from expectation (by about 15%), the latest measurement is both much more precise (0.4%) and in agreement with theoretical expectation [260]. Meanwhile studies have started on a second isotope, $^{207}\text{Pb}^{81+}$, that required additional experimental tricks [220]. The lifetime value extracted is not (yet) as precise as the above.

5. Prospects

One of the physical problems associated with the topic of this review is the measurement of (not so long) atomic lifetimes (low transition rates) in neutral heavy atoms. On parity-nonconservation (PNC) measurements in Cs, done by observation of an M1 transition in Cs, Bennett and Wieman [24] conclude that “In all cases, the new measurements show better agreement with the calculations than earlier measurements and also show that the large previous discrepancies were likely due to experimental errors.”

For the intercombination and forbidden transition rates discussed here, much closer to classical physics, the data presented above tell a different story: While certainly some experiments were evaluated with results that no longer hold, theory yielded a similar scatter of lifetime numbers. Considerable development of both experiment and theory was needed to constrain the ranges of valid data by a factor of ten or more. In hindsight, some old predictions turn out to have been close to later knowledge, but they often carried considerable uncertainty (without any indication to its magnitude). The new storage ring results on intercombination and forbidden transition rates in few-electron ions are all more precise than present calculations, even after semi-empirical corrections of the latter, and they thus provide a long-missed challenge to theory.

The above claim about PNC theory and experiment reminds me of the situation 20 years ago, when precision wavelength measurements by the beam-foil technique, on two-electron-ions, suddenly advanced to a precision that would be testing quantum electrodynamics (QED) at the level of better than 10%. The renewed theoretical efforts then resulted in an improved precision of QED predictions. The new results, however, are hard to test experimentally, because the dominant uncertainty of the theoretical atomic structure values results lies in the non-QED parts of

relativistic quantum mechanics. Instead of testing QED on the basis of known classical physics and quantum mechanics, nowadays the QED part is largely taken for granted and the testing applied to the non-QED fundamentals. It is, of course, quite possible that a similar story will evolve about PNC, revealing the insufficiency of knowledge and detailed understanding of the non-PNC wave functions that are here to be tested by lifetime measurements.

Obtaining any data in first place was a challenge for experiment and theory. Producing reliable and fairly precise data – as presented by the data in this review – was an important step forward. What is next, is less fun for the people involved, in theory and experiment, and certainly less cherished by peer reviewers working for funding agencies and journals: The improved experimental and calculational techniques need to be extended to other atomic systems, producing no foreseeable fancy *new physics*, but valuable data and reliable material for cross checks of experiment and theory in more complex systems. Moreover, the next order of magnitude of precision lifetime determinations deserves to be tried. Over and over again in physics, the strife for higher precision has illuminated the limits of knowledge on old and demonstrated the existence of new phenomena. The further insight to be gained from even better atomic lifetime measurements using ion traps of all sizes certainly is valuable for this quest.

Conventional ion traps, with electron ionization and excitation of the ion species of interest from some atomic or molecular gas, or from a laser-produced vapour plume [145,163] have served physics well, but have many problems (most notably the vacuum working range) that preclude the gathering of truly precise lifetime data. Since meanwhile other techniques have become available for that purpose, it would seem wise to use conventional traps for lifetime experiments on molecules rather than for further run-of-the-mill atomic ions [48,176,177]. For molecules the ease of production and flexibility of operation are rather advantageous, and the attainable experimental and theoretical precisions match nicely. Of course, heavy-ion storage rings are also used for lifetime measurements on molecules, negatively and positively charged ones, [3,4], and the small storage ring ELISA at Aarhus has been set up for exactly such studies.

There presently are two experimental set-ups that combine an ECR ion source with a Kingdon trap, Church’s old set-up that is now being run at the Jet Propulsion Laboratory, and the newer set-up at the University of Nevada Reno. The rate at which the latter are claimed to produce data is amazing (the statement in a recent paper is “two hours per good decay curve” [191]). At this speed, most of the forbidden transitions listed in the Kaufman/Sugar tables [149] seem in reach within less than a year. However, I have heard with sorrow that the funding of this project has ended. I also note that the publications actually show only a few cases that resulted in such precise lifetime results, even for ions like those of Mn that the ECR ion source appears to produce easily. Last, but not least, I have the impression (fed by comparisons of their results with those from EBIT – that also are far from perfect so far – and from the possibly more reliable MeV-ion beam experiments at a heavy-ion storage ring [243]) that the systematic uncertainties of the Kingdon trap lifetime measure-

ments may be larger than the authors realized. More work and even more careful investigations will be necessary to ascertain that the high level of precision claimed for some of the data is correct.

EBIT also has problems when aiming at high precision lifetime data. At high electron beam energies, the electron current maybe high and result in a high signal rate, but also in the production of higher charge-state ions and in contributions from recombination, which may corrupt the high data quality required [242]. Precision lifetime experiments will need to explore further the parameter space for hidden obstacles. Almost certainly they will be limited to electron beam energies below the excitation threshold of the next higher charge state ion, where the experimental conditions are cleaner than otherwise, but where the electron beam current is far from maximum and thus also the signal rate (in a recent run this amounted to one order of magnitude) (Fig. 3). However, a precision of lifetime data at the 0.1% level in X-ray work, and at the 1% level for lifetime measurements in the visible, seems within reach [240,242]. Most recent EBIT lifetime work on visible lines at LLNL [22], studying various systematic effects, shows that such high precision is, indeed, attainable in the visible spectral range and that the first experimental lifetime results of this quality fall into the same isoelectronic trends as the storage-ring data – which are the ones least troubled by systematic error. In fact, these data disagree with some of the Kingdon trap results (and, naturally, I prefer to trust my own – storage ring and LLNL EBIT – experiments and error estimates).

EBIT has the enormous advantage of being applicable to many elements, and in particular to high charge states. For some of the hyperfine-induced decays in (effective) two-electron ions, I see EBIT as the presently only way open. Furthermore, EBIT will be essential to identify various forbidden lines that appear in the VUV and EUV spectrum of the solar corona. EBIT can be used to establish the elemental species and the ionization energy of the ions that emit these lines, and it will be (fairly) straightforward to obtain decay curves, too. The lifetimes, which have been calculated only recently [161] for an ion that is abundant in the solar corona, Fe^{9+} , and not yet for many others, will be useful for modeling the processes in the solar corona. Why not jump from no available measurement directly to precise lifetime measurements – as far as possible – and challenge calculations even more?

The heavy-ion storage ring is the most promising tool for precision lifetime measurements, because it combines isotope and charge state selectivity with very good vacuum conditions and easy control of the stored ions and their storage behaviour. Lifetime error margins of 0.2% and better have been reached by several experimental techniques at TSR, and further improvements of this precision are quite feasible (though not exactly cheap in terms of equipment and accelerator time).

Measurements on low-charge state rare gas ions (like Ne^{2+} , $\text{Ar}^{2+,3+}$) ought to be done at a storage ring in order to check on the RF trap data on such ions that are present in many technical plasmas. At TSR, these particular experiments will require the use of a single-stage injector, since the rare gases do not feature the negative ions needed for the tandem accelerator employed so far.

The present heavy-ion storage rings have been designed for the extended storage of ions with a high charge-to-mass ratio, but apparently they also do very well for lifetime measurements on low-charge state light ions. TSR, for example, can be developed for lifetime measurements of higher-charge state heavy ions by implementing the presently planned high-charge state ion injector (based on an RFQ system) which is to be completed by an ECR ion source. Then an interesting project will be the study of extended isoelectronic sequences, a massive improvement on the statistical quality of the aforementioned data on highly charged Ti ions (B- and F-like), as well as a quest to obtain data on similar ions of Ar – enabling a direct comparison to the Kingdon trap and EBIT data on these ions that also appear in the solar spectrum. By coupling to high-energy injectors, like GSI's ESR that is fed from the SIS synchrotron, heavy-ion storage rings can even reach arbitrarily high charge states with sufficient intensity. However, excitation then has to be achieved in a device like GSI's fragment separator FRS or (nondestructively?) inside the storage ring, as has been demonstrated by using laser-excitation on H-like Bi^{82+} and Pb^{81+} , or by single electron capture in the cooler or in an internal gas target. Applied to the physical effects to be studied in such high- Z systems, lifetime data of a precision at a much coarser level than reached for some low- Z systems may already render quite meaningful tests of specific calculations. However, on a grander view, the challenge lies in getting closer to a detailed understanding of neutral heavy atoms, and for this the precise measurement of atomic transition rates in *low* ionic charge states is of particular interest. Thanks to the various ion traps, the first meaningful steps on this road have been taken.

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