REVIEW/SYNTHÈSE

Precise atomic lifetime measurements with stored ion beams and ion traps

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Abstract: For many years, atomic lifetime measurements on multiply-charged ions have been done almost exclusively by beam-foil spectroscopy. For low ion charges, however, spin-changing "intercombination" transitions have a rate that renders them too slow for traditional fast-beam techniques. Here ion traps and fast-ion beams have been combined in the concept of heavy-ion storage rings. These devices have permitted not only an extension of intercombination lifetime measurements down to singly charged ions, but they also facilitated similar measurements on electric-dipole forbidden transitions. The electron-beam ion trap (EBIT) complements the storage-ring work for work on highly charged ions. Achievements, technical issues, and prospects are outlined.

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Résumé : Depuis plusieurs années, les mesures de durée de vie dans les ions multi-chargés ont été effectuées presque exclusivement par la spectroscopie faisceau-lame (« beam-foil »). Cependant, pour les ions faiblement chargés, les transitions « inter-système » qui impliquent un changement de spin sont trop lentes pour permettre leur mesure par des techniques conventionnelles qui utilisent les faisceaux rapides. Dans de tels cas, les techniques des faisceaux d'ions rapides et des pièges ioniques ont été combinés dans les anneaux de stockage d'ions lourds. Grace à ces dispositifs on a pu mesurer des durées de vie qui impliquent des transitions inter-système allant jusqu'aux ions simplement chargés ainsi que des transitions interdites par les règles de sélection dipolaires électriques. Le piège ionique faisceau d'électrons (« EBIT : electron-beam ion trap ») est une technique complémentaire pour les ions fortement chargés. Les résultats obtenus jusqu'à date, des aspects techniques ainsi que des perspectives d'avenir seront discutés.

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Dedicated to Professor Indrek Martinson at the occasion of his 65th anniversary.

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1. Introduction

Atomic lifetime measurements of electric-dipole transitions yield information on atomic wave functions that supplements the data from atomic energy levels, but is more sensitive to details of the radial wave functions. This is evident as the electric-dipole operator, er, used in describing the most common electric-dipole transitions, (E1), in atoms itself depends on r. Transitions between fine-structure levels of a given term are supposedly insensitive to this, as they connect levels with similar radial wave functions. However, complex wave functions as well as relativistic effects in highly charged ions modify this simple picture.

Transition rates of intercombination (spin-changing) and electric-dipole forbidden transitions are of interest not only for the detailed understanding of atomic structure and dynamics, but also for applications in plasma diagnostics and astrophysics. The relative intensities of resonance and intercombination transitions are often used as a tool for density diagnostics, if the collision rates in the plasma are in between the radiative rates for these particular transitions. The low transition probability of some intercombination and many forbidden transitions gives them an advantageous larger optical depth in astrophysical observations. Forbidden lines in the ground-state configuration of highly charged ions can often be found in the visible spectrum (like the solar coronal lines) and thus are amenable to high-resolution spectroscopy and line diagnostics, while most other prominent lines in the spectra of the same ions lie in the vacuum or extreme ultraviolet (VUV/EUV) ranges, for which high spectral resolution is much more difficult to achieve or direct observation from the ground (for solar spectra) is not even possible.

Until fairly recently, the measurement of intercombination transition rates in low-charge-state ions and of electric-dipole "forbidden" fine-structure transition rates had not yet been achieved, and evaluations of terrestrial and astrophysical plasma spectra that used "line ratios" had to rely on calculations only. However, in particular, for forbidden transitions between fine-structure levels of a given term, theory was considered adequate if not outright simple, since in the nonrelativistic limit and single-configuration approximation the problem appeared to be reducible to Racah algebra and measured energy splittings [1]. The results of some of the measurements to be discussed here clearly challenge this view. Experiments at a heavy-ion storage ring have determined intercombination transition rates in various low-charge ions to a precision better than 1%, an improvement on typical radio-frequency (RF) or electrostatic ion-trap measurements by typically an order of magnitude. In this, they are regularly surpassing the theoretical precision, and in some cases they call for a revision of accepted calculations. Similar measurements on forbidden transitions suddenly change the situation from a "theory-only" to a "theory-tested-and-repeatedly-found-lacking" one. The new measurements often are much more precise than would be needed for any practical applications. However, in the evaluation of many astrophysical observations, well-known atomic parameters will permit re-evaluation of the error budgets. Moreover, precise, accurate atomic data provide benchmarks for theory. Calculations will be required to obtain numbers in all those cases of interest that are not accessible to experiment for whatever reason, and theory better demonstrates its validity for the benchmark systems before reliability claims are made for the more complicated cases.

For a long time, beam-foil spectroscopy has been the only technique available that permitted the measurement of the atomic lifetimes of ions of any element and in any charge state [2]. In this technique, a beam of fast ions is sent through a thin foil in which it suffers collisions mostly with the electrons of the foil material. The ions loose a small fraction of their energy, but still emerge as a rather well-defined beam of ions that have little spread around the mean velocity. The mean charge state and the charge state distribution of the ions leaving the solid depend on the ion energy. At energies of order 100 keV, the ion beam may contain a negative-charge state fraction; at 500 MeV/nucleon even U can be stripped of all its electrons. (The same ion–electron collision energy is available in collisions of 250 keV electrons with atoms at rest — a fact exploited in the electron-beam ion trap [3–6] discussed below.) Displacing the foil from the line of sight of the spectrometer permits the recording of light at given times after excitation,

and thus of decay curves (from which atomic lifetimes can be extracted) or of delayed spectra [7]. Several reviews (see, for example, refs. 8–11) have tracked the progress of this technique.

Lifetime measurements on the magnetic dipole (M1) decay of the $1s2s\ ^3S_1$ level in He-like ions (by various techniques) now reach from neutral He atoms to Xe^{52+} ions (with almost all data for $Z > 16$ obtained by beam-foil spectroscopy, see ref. 12 and references cited therein); the intercombination transition rate ($2s^2\ ^1S_0$–$2s2p\ ^3P^o_1$) in Be-like ions has been measured from B^+ to Xe^{50+} (see ref. 13); the resonance line in Na-like ions and intercombination transition rates in Mg- and Al-like ions have been measured up to Au ($Z = 79$) [14–16]. The limits of the accessible lifetime range are given by the spatial resolution of the detection system (which for beam-foil spectroscopy corresponds to time resolution) for short lifetimes (a few picoseconds are the usual lower limit), and by the ion-beam optics and experimental chamber size for long lifetimes (30 ns or 1 m chamber lengths are rarely exceeded). For longer lifetimes, the decay curves are spread out so much along the ion beam that the signal level obtained with any regular-sized detector is insufficient. There are two ways out of this dilemma: Either bend the ion beam around and let it circulate in an ion trap, be it small (10 cm diameter electrostatic (Kingdon-type) trap for keV-ions [17]) or large (50 to 100 m circumference heavy-ion storage ring for MeV to multi-MeV ions), or produce ions of low kinetic energy inside a trap and switch off the production mechanism (for example, the electron beam in the electron-beam ion trap) while watching the decay.

Ion traps of various types (electrostatic (Kingdon), magnetic (Penning), and radio frequency (Paul)) have been around for quite some time (for a comprehensive book on the subject, see ref. 18, for an earlier review on atomic lifetime measurements using ion traps, see ref. 19, for a more recent one see ref. 20) — what is new? Lifetime measurements on trapped ions in low-charge states ($q = 1$–3) have been done in the lifetime range from a fraction of a millisecond to a few seconds, but until recently the precision rarely reached 5%. The new results are in an improvement in measurement precision of more than an order of magnitude, reducing statistical and systematic errors, and the inclusion of highly charged ions. This gain in accuracy, precision, and measurement range is much more than just another incremental step of technical progress, more than correcting some blunders in earlier work and gradually improving the data base. In highly charged ions, relativistic effects are important if not dominant. However, the overwhelming term in the Hamiltonian operator is the nuclear Coulomb field, a well-defined central potential. Therefore, calculations for highly charged ions converge much better than those for low-charge ions or neutral atoms, and experiments on highly charged ions can strive to test calculations that seem firmly grounded. Near the neutral end of iso-electronic sequences, in contrast, electron correlation is important. Treating it well requires wave functions with very extensive basis sets consisting of tens of thousands of components. Relativistic approximations are automatically even more complex, but are fundamentally more consistent. Evidently, the degree of complexity grows towards the neutral end of any iso-electronic sequence. That is not only the range closer to our everyday world of neutral atoms and a few singly charged ions, but also the range in which extremely sensitive experiments probe parity violation or in which ions are prepared in traps for improved ultra-stable frequency standards. Hence, this is the direction towards which all calculational and experimental efforts ultimately aim. At present, some of the experiments on low-charge ions included in this review are ahead of theory in terms of precision.

What has hindered ion-trap measurements from reaching the new precision and ion-charge ranges earlier? The most frequently used method to catch ions in a trap started out by producing the ions inside the trap, regularly by electron bombardment of a gas or vapour. At a higher gas pressure one makes more ions, but these ions then also suffer charge-changing and other detrimental collisions with the residual gas at a higher rate, and high-charge-state ions are not accessible in sufficient quantity this way. Lifetime measurements were done at various pressures (often in the 10^{-7}–10^{-8} mbar range) and the results extrapolated to zero pressure. The data closest to the clean conditions (at zero pressure) are implicitly also the ones with the lowest signal rate, which keeps the uncertainty of the extrapolation high.

The new techniques and devices mentioned here all work better in a vacuum ($p < 10^{-9}$ mbar for the electrostatic ion trap at Reno, $p < 10^{-10}$ mbar for the electron-beam ion trap and the heavy-ion storage ring), ion production is often external to the trap (mass spectrometry and spectroscopic experiments on radioactive ions use corresponding techniques and separate, specifically designed traps for ion transfer and ion storage), and a preselection of charge states is possible. The newly accessible range of higher charge states opens up to measurement some atomic systems that simply are too long-lived in low-charge-state ions. Various combinations of these factors have been realized and will be discussed in the following.

2. Ion-trap experiments

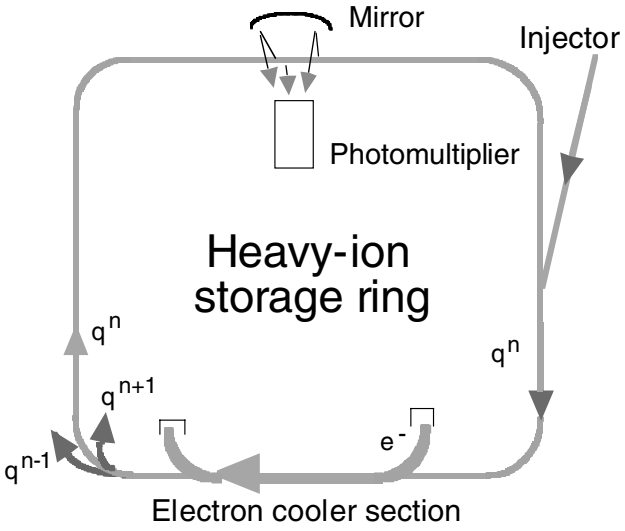
2.1. ECR ion source with Kingdon-type ion trap

An example of the decoupling of ion production and trapping for atomic lifetime measurements has been set by capturing ions from an ion beam passing across the volume of an electrostatic ion trap [17, 21]. The ion beam is provided by an electron cyclotron resonance ion source (ECRIS) and is charge-selected by a magnet in the beam transport system. When the voltages on the trap are suddenly raised to working conditions, a section of the beam (of, say, 10 cm length) is caught inside. Not all of the ions have trajectories that are compatible with lasting capture, but about half of them settle into an ion cloud that lasts for a few seconds, while optical observation can take place. Then the leftover ions are ejected (by a voltage pulse), and some of them hit a detector that serves as a monitor. From ion ejections after different trapping times the typical loss rate can be established, which is needed to correct the optical decay curves. Recent measurements on Ar, Mn, and Fe ions yielded atomic lifetimes in the range of a few to many milliseconds, and an ion storage-time constant of around 1 s has been mentioned [22–24]. (Whether the ion survival measurement in the midplane of the trap is truly representative of the full ion cloud that is observed optically remains an open question.)

Two such systems exist now; an older one previously used at Texas A&M has been moved to the Jet Propulsion Laboratory (JPL) at Caltech, while a newer one, constructed by the same group, has been set up at Reno (Nevada) [17]. The ECR ion source in this arrangement is advantageous for the production of ions in all technically or astrophysically interesting charge states (reported data reach up to $q = 14^+$). Uncertainties of atomic lifetimes have been reported in the range 1–15% in the most recent round of experiments [22–24]. It should be noted that the ion energies in these experiments are in range of a few keV. This is better for clean experiments than the eV energies in conventional ion traps with their in situ ion production, but the collision cross sections are still high, compared to the MeV energies used with storage rings. While theory may deserve new efforts now that it can be tested, some deviations between the results from these Kingdon-trap experiments and recent theory are surprisingly large. In contrast, almost concurrent experiments at the Heidelberg heavy-ion storage-ring TSR and at the Livermore electron-beam ion traps EBIT-I and EBIT-II — which treat some of the same cases — so far come out in much better agreement with theory (see discussion below). In this comparison, it seems likely that in the electrostatic ion-trap work with highly charged ions (in particular the partly pioneering work using the Kingdon-trap experiment at Reno [17, 22–27]) the influence of systematic error may have been underestimated. Apparently, the experimental set-up at Reno is no longer in active use, which makes it difficult to check out various suggestions of sources of systematic error with the original equipment.

A new design for an electrostatic trap has been developed in Israel by Zajfman and his colleagues. It uses injection along the axis of a 40 cm long tube and reflects the ion beam from the far end before the near-end cap voltages are raised to close the trap. The trap has been used with various singly charged ions (both positive [28, 29] and negative [30]) of energy 4.2 keV, as well as with Xe^{2+} ions [31]. When ions are neutralized (or drastically reduced in charge state) they can leave the trap through the electrostatic barriers at the ends and are then detected by a microchannel plate. A related device of striking simplicity in design, called the cone trap, is being developed by Cederquist and his colleagues at Stockholm [32].

Fig. 1. Schematics of a heavy-ion storage ring with particle detection (bottom) and photon detection (top). To indicate the scale: the Heidelberg heavy-ion storage-ring circumference is 55 m, while the photomultiplier tube has a diameter of about 25 mm.



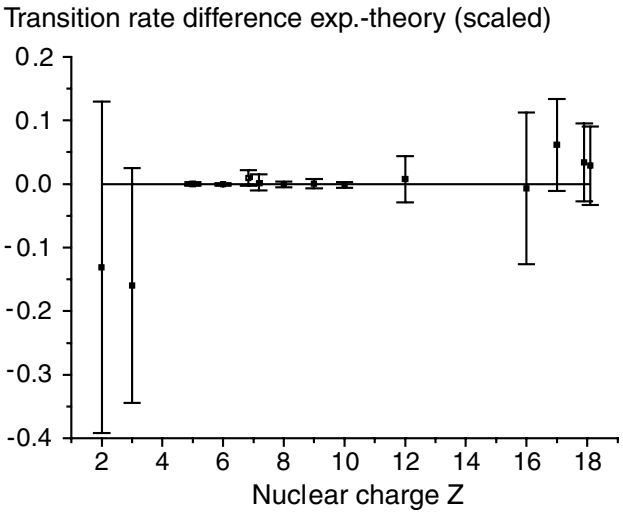
Similar work on the autodetachment lifetimes of negative ions has been done extensively at the Aarhus storage rings ASTRID [33, 34] and ELISA. Owing to the developing competition in the field, there now are cross checks available of results from different laboratories. For example, the precision of lifetime results on Be^- obtained at the magnetic storage ring ASTRID at Aarhus has been improved upon using the electrostatic ion trap in Israel; yet more recent experiments at the new electrostatic storage ring ELISA at Aarhus have achieved an even higher precision [35] and also claim better accuracy than obtained with the electrostatic ion trap. However, autodetachment of negative ions is a field of its own; this review concentrates on radiative lifetimes of levels in positive ions.

3. Heavy-ion storage ring

The ultimately long trap to capture a section of an ion beam is a ring vessel in which to thread the beam (Fig. 1), which is exactly the working scheme of a heavy-ion storage ring [36, 37]. As with yarn on a spool, the freshly injected ion beam in the ring is slightly displaced after a turn so as to accept the next turn. This technique, called stacking, permits the accumulation of a stored ion beam over about 30 turns (at the test storage ring (TSR) Heidelberg) before the phase space of the ring is filled and injection ends; the beam can then be left coasting for minutes. 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 permits the addition of more such accumulation cycles. Such a cooled ion beam is of great interest for many detailed atomic physics studies. For low-charge-state ions, however, cooling times of order 1 s and more are too long in comparison to the atomic lifetimes of interest, and no advantage would be gained from such cooling.

Storage rings for atomic physics so far are a European specialty, with atomic lifetimes being reported from ASTRID and ELISA (Aarhus), CRYRING (Stockholm), ESR (Darmstadt), and TSR (Heidelberg). These rings have some very different parameters as far as ion sources, ion energies, and ring size are concerned. Similarly, the available and applicable detection techniques differ among the laboratories. Particle detection has been used to measure atomic lifetimes that relate to the autodetachment of the "surplus" electron of negative ions [33, 34]. It was also employed to detect dielectronic recombination (DR) of stored ions with electrons in the electron cooler as a probe of excited level populations [38, 39], or simple ionization (effected by the cooler electrons or by collisions with the residual gas) [40]. All

Fig. 2. 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 low-Z range $Z \leq 18$ is shown. The particularly precise data for ions in the range $5 \leq Z \leq 10$ underline the motivation for the present review: precise data are just so much more meaningful. Data for $Z = 5$ to 7 (leftmost entry) are from the heavy-ion storage ring TSR [38,39], exploiting dielectronic recombination processes for monitoring the excited level population. The other data for $Z = 7$ to 12 have been obtained by way of X-ray observations at the Livermore electron-beam ion trap EBIT-II [41–43]. All experimental data have been scaled by normalization to the results of the fully relativistic calculation by Johnson et al. [44], and only the deviations from this prediction are displayed. For a discussion of these particular measurements and for a comparison to other calculations, see ref. 43.



of the experiments with particle detection rely on the fact that the storage-ring parameters (magnetic fields) are optimized for a given charge-state ion species. Ions that have grabbed an electron and thus reduced their electric charge will be less deflected than the majority species by the next bending magnet (neutralized ions even fly on straight), and, therefore, they can be detected on the outside of the ring. Ions that suffered further ionization can be intercepted on the inside of the track (Fig. 1). The DR process has resonances so that a state-specific detection process is possible, and it works very well for two-electron ions [38, 39]. In experiments aiming at the $1s2s\ ^3S_1$ level in the He-like ions B^{3+}, C^{4+}, and N^{5+}, a lifetime measurement precision of as low as 0.2% has been achieved (Fig. 2). However, experience with four-electron ions [40] suggests that the variety of atomic systems that are suitable for the DR technique may be rather limited (because of the low production efficiency and other complications), even as the detection efficiency for particles is high.

Instead, optical techniques (usually of low detection efficiency, of order 10^{-7} overall) have been tried and were found to be exceptionally helpful, yielding precise data [45]. MeV-ion beams in storage rings can last for many seconds, minutes, sometimes hours, because the vacuum is very good and the collision cross sections at these energies are very small. However, some time (less than 100 μs) is needed for beam transport from the ion source and the accelerator to the storage ring (and for a number of turns in there, each of order 10 μs). From this, the optimum working range for atomic lifetime measurements can be estimated to reach from a few hundred microseconds to a few seconds. Experiments so far exploit the excitation of long-lived or metastable levels in the ion source or the stripper of the injector accelerator. Excitation (also after cooling) inside the storage ring faces major difficulties, as interactions with solid matter would be destructive for the ion beam. Consequently, excitation by collisions with a gas target or with the electrons in the cooler section has been employed for spectroscopy, but not for atomic lifetime measurements, since ions after a successful charge change would no longer be stored.

There also are experiments that use laser light to probe the level population of a stored ion beam

or to shift it from a metastable level to a short-lived level, the decay of which can then be readily observed [46–48]. However, the light from present-day lasers matches only few transitions in highly charged ions, and none that would excite a highly charged ion from a low-lying level to any high-lying one. For this reason, the atomic lifetime experiments at the Stockholm storage ring concentrate on singly charged ions (which offer a plethora of interesting cases, in particular in the astrophysical context). However, there are plans for exposing an electron-beam ion trap to X rays from the (yet-to-be-built) free-electron laser at the TESLA test facility at Hamburg.

In all ion-trap lifetime measurements, it is of paramount importance to determine the ion storage time (the inverse of the loss rate). The observed optical signal decay rates represent the sum of the true atomic-decay rate and of all other loss processes, notably the loss of ions from the stored sample. With the single-charge-state ions stored in the storage ring, this measurement is relatively straightforward, measuring rest gas ionization, monitoring the ion beam current via a transformer coil, laser-probing other levels of the ion sample, etc.

3.1. Electron-beam ion trap

Electron-beam ion traps 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 50 μm. The "Penning" parts of the trap are completed by drift tubes at 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 (for example, a neutral gas stream of a density corresponding to a pressure of as low as 10^{-10} mbar may be crossing its trajectory under UHV conditions ($< 10^{-11}$ mbar)), which are then confined by the trap fields and can be hit over and over again. If the electron–ion 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 determines the need for UHV). The charge-state 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. For lifetime measurements, the electron-beam energy is modulated above and below the production and (or) excitation threshold [49], or even shut off completely after a period of ion production. This then is the magnetic trapping mode [50] in which EBIT acts as a Penning trap with ion storage times of many seconds. The ion cloud does expand somewhat when the electron beam is switched off, but trapping is still effective for many seconds (as has been ascertained by ion-cyclotron resonance-frequency observations) [50, 51].

However, measuring the trapping behaviour of a given ion species in the context of lifetime measurements can be difficult. One of the preferred techniques of monitoring the presence of ions in the electron-beam ion trap is by detecting their X-ray emission. However, there usually is no X-ray emission at the times after excitation (when the electron beam is switched off) that are important for optical lifetime measurements on long-lived levels, except from charge-exchange events (collisions with the rest-gas atoms), and those are few and far between. Moreover, ions in many of the charge states of interest in various plasma physics or astrophysical applications do not radiate in the X-ray range at all, because all inner shells are filled. Then one may resort to measuring the trapping times of more highly charged ions instead (which are easily produced by increasing the electron-beam energy), but, of course, this supplemental choice carries its own uncertainties. Hence, it is evident that trapping times can be conveniently long, but it may be difficult to actually specify a trapping time value for a given ion species in a given situation [52].

Table 1. Most precise results of the ion-trap lifetime measurements on ions with intercombination transitions (E1, $\Delta S = 1$).

Ion	Level	Lifetime τ	Trap type	Ref.
Be sequence				
B^+	$2s2p\ ^3P^o_1$	(97.65±0.5) ms	HSR	53
C^{2+}	$2s2p\ ^3P^o_1$	(9.714±0.013) ms	HSR	45
O^{4+}	$2s2p\ ^3P^o_1$	(0.458±0.015) ms	HSR	54
B sequence				
C^+	$2s2p^2\ ^4P^o_J$	(7.95±0.07/104.1± 0.5/22.05±0.07) ms	HSR	56
N^{2+}	$2s2p^2\ ^4P^o_J$	(1.32±0.05/13.9± 0.2/3.3±0.07) ms	HSR	56
C sequence				
N^+	$2s2p^3\ ^5S^o_2$	(5.88±0.03) ms	HSR	57
O^{2+}	$2s2p^3\ ^5S^o_2$	(1.22±0.08) ms	RFT	58
		(1.25±0.02) ms	HSR	59
Mg sequence				
Al^+	$3s3p\ ^3P^o_1$	(0.300±0.01) ms	RFT	60
		(0.305±0.010) ms	HSR	53
Si^{2+}	$3s3p\ ^3P^o_1$	(59.9±1.8) μs	RFT	61

HSR, heavy-ion storage ring; RFT, RF ion trap.

4. Experimental data

4.1. Intercombination transitions

The simplest atomic systems that feature millisecond lifetimes and decay branches in the UV part of the spectrum are the four-electron ions (Be iso-electronic sequence), with the $2s^2\ ^1S_0$–$2s2p\ ^3P^o_1$ intercombination transition. Taking an example from the ESR ring at Darmstadt, measurements at the Heidelberg TSR use a light-collecting mirror in the ring vessel and a solar-blind photomultiplier outside a sapphire window, viewing a 5 cm long section of the stored ion beam [45]. Data acquisition is started in synchronicity with the ion-storage cycle. There are no mechanical movements, and the corrections to the photon signal consist of only the ion-loss rate (a very small correction for atomic lifetimes in the millisecond range and with storage time constants in the many-second range) and relativistic time dilation. Transient-field effects that might mix fine-structure levels have been estimated to have no notable influence in the cases studied so far.

Early calculations of the rate of this transition in C^{2+} scattered by about 20% around the mean, 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 recently obtained storage-ring data on C^{2+} (uncertainty ±0.13%) [45], B^+ (±0.5%) [53], and O^{4+} (3%) [54] agree with some of the latest theoretical results from massively improved calculations (see ref. 13), but are more precise than most of them. There is slight disagreement, however, with the first calculation for C^{2+} that claims an uncertainty as low as 0.5% [55]. All of these recent calculations employ different basis sets for initial and final states to account for the relaxation of the electron-density distribution and other subtle effects. While the lifetimes of the short-lived resonance levels in Be-like ions apparently can be calculated to better than 10^{-3} — but not measured to anywhere near this precision — the intercombination rate can now be measured to a fraction of 1%, but cannot regularly be calculated equally well. The presently available precise ion-trap lifetime data on such intercombination transitions are summarized in Table 1. (A tabulation that includes additional less precise data may be found in a recent review [20].) The above case of C^{2+} is not only interesting as an exercise in precision, but also of interest for astrophysics [62]. Carbon is one of the more abundant elements in the Universe, and while much of the (in principle) visible matter of the Universe is in the plasma state, most ions are in low-charge states. In the Be

sequence, the neighbouring level to the level discussed above, $2s2p\ ^3P^o_1$, is the $2s2p\ ^3P^o_2$ level, with a magnetic quadrupole (M2) decay branch to the ground state, $2s^2\ ^1S_0$, and an M1 decay branch to the $^3P^o_1$ level. For high-charge states of Be-like ions the M1 decay branch dominates, but for low-charge-state ions (like C^{2+}) the M2 transition is the dominant one. Thus, there are two spectral lines, from the ground-state decays of the levels $2s2p\ ^3P^o_1$ and $2s2p\ ^3P^o_2$, that are close together in wavelength, but still easily resolved. In C^{2+}, one of the levels has a lifetime of near 10 ms (this is the aforementioned one that has been measured at the storage ring [45]), while the other has a predicted lifetime of about 180 s [13] (which is too long for present storage and measurement techniques). The intensity ratio of these two lines is then sensitive to the (very low) particle density of some far-away interstellar clouds. There has been discussion that there might be a third line nearby, the hyperfine-induced decay of the $2s2p\ ^3P^o_0$ level in the isotope ^{13}C, but apparently this line of very low transition probability, and from a low-abundance isotope, has not yet been observed.

The precision of the C^{2+} lifetime data obtained at the heavy-ion storage ring corresponds to that of the best fast-beam laser work on neutral alkali atoms. While theory matches experiment in precision for the light alkalis, there are severe discrepancies (of order 20%) for the heavy alkali atoms [63]. Similar problems may be occurring with 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). The Mg iso-electronic sequence features the same valence shell level structure (for $n = 3$) as the Be sequence does (for $n = 2$), and the problems of calculating the $ns^2\ ^1S_0$–$nsnp\ ^3P^o_1$ intercombination transition rate of electrons outside a core of closed shells might be assumed to be not very different for either $n = 3$ or $n = 2$. Interestingly, the measured sub-ms level lifetime in Al^+, with RF ion trap [60] and heavy-ion storage-ring [53] experiments yielding practically identical results, is not in good agreement with recent calculations.

A different complication arises in the C iso-electronic sequence. The decay of the $2s2p^3\ ^5S^o_2$ 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. The storage-ring results (5.88 ± 0.03 ms [57]) with their much higher precision (0.5%) lie just outside the 5% (1σ) error bars of both of the latest experiments (using an electrostatic ion trap) and calculations. The calculational problems relate not just to multiplet mixing (because of spin–orbit interaction), but to severe cancellation effects in the transition matrix elements of the mixing partners in the triplet-term system. Meanwhile, renewed theoretical efforts [64, 65] tend towards the most precise experimental number. A very recent storage-ring measurement on the same level in O^{2+} [59] corroborates the earlier RF ion-trap results [58], but reaches a higher precision. Similarly, it deviates slightly (and with the same sign) from results obtained with the same type of earlier calculation as was used for N^+.

A challenge to the heavy-ion storage-ring technique is the three-component decay curve of the $2s^22p\ ^2P^o_{1/2,3/2}$–$2s2p^2\ ^4P_{1/2,3/2,5/2}$ multiplet in the B-like ions C^+ and N^{2+}, with predicted lifetimes (for C^+) that range from 5 to 100 ms. Earlier RF ion-trap data were at variance with most calculations, as the pattern of the three lifetimes did not fit any of them well. Such an experiment needs to obtain data of very high statistical significance so that the extraction of three exponential components with reliable parameters becomes possible, from a common, spectroscopically blended, decay curve. This has, indeed, been achieved in a recent storage-ring experiment [56], and the lifetime pattern found does match that of several — not so recent — calculations. Another recent re-measurement of these same decays in C^+ [66] used the Kingdon-type ion trap at Caltech, but fell somewhat short of the statistical quality of the new heavy-ion storage-ring data.

4.2. Forbidden transitions

Forbidden transitions, that is, transitions within an electron configuration that are forbidden for electric dipole transitions because parity does not change, have been found as the origin of the solar corona lines in the visible spectrum. The identification of such lines by Edlén [67] with fine-structure transitions in highly charged Ca and Fe ions solved a riddle that was many decades old. In turn it

Table 2. Examples of recent precise experimental lifetime results on ions with M1 and (or) E2 transitions. Where several results are available, the most reliable (in the present author's view) was selected.

Ion	Level	Lifetime τ	Trap type	Ref .
He sequence				
B^{3+}	$1s2s\ ^3S_1$	(149.8±0.45) ms	HSR	39
C^{4+}	$1s2s\ ^3S_1$	(20.63±0.05) ms	HSR	38
		(20.589±0.042) ms	HSR	39
N^{5+}	$1s2s\ ^3S_1$	(3.905±0.05) ms	HSR	38
		(3.94±0.05) ms	LLNL EBIT	43
O^{6+}	$1s2s\ ^3S_1$	(956±5) μs	LLNL EBIT	41
F^{7+}	$1s2s\ ^3S_1$	(274±3) μs	LLNL EBIT	43
Ne^{8+}	$1s2s\ ^3S_1$	(91.7±0.4) μs	LLNL EBIT	42
Be sequence				
Ar^{14+}	$2s2p\ ^3P^o_2$	(15.0±0.7) ms	Oxford EBIT	72
		(15.0±0.8) ms	LLNL EBIT	73
B sequence				
Ar^{13+}	$2s^22p\ ^2P^o_{3/2}$	(9.70±0.15) ms	LLNL EBIT	73
Ti^{17+}	$2s^22p\ ^2P^o_{3/2}$	(0.627±0.010) ms	HSR	74
C sequence				
O^{2+}	$2s^22p^2\ ^1S_0$	(530±25) ms	HSR	59
F^{3+}	$2s^22p^2\ ^1S_0$	(304±5) ms	HSR	59
Si^{8+}	$2s^22p^2\ ^1D_2$	(38.3±0.3) ms	HSR	75
N sequence				
S^{9+}	$2s^22p^3\ ^2P^o_{1/2}$	(5.0±0.25) ms	HSR	59
S^{9+}	$2s^22p^3\ ^2P^o_{3/2}$	(2.0±0.2) ms	HSR	59
O sequence				
F^{+}	$2s^22p^4\ ^1S_0$	(420±12) ms	HSR	76
Si^{6+}	$2s^22p^4\ ^1D_2$	(63.6±0.7) ms	HSR	75
F sequence				
Ar^{9+}	$2s^22p^5\ ^2P^o_{1/2}$	(9.32±0.12) ms	LLNL EBIT	73
Sc^{12+}	$2s^22p^5\ ^2P^o_{1/2}$	(1.00±0.03) ms	HSR	53
Ti^{13+}	$2s^22p^5\ ^2P^o_{1/2}$	(0.513±0.010) ms	HSR	74
Al sequence				
Fe^{13+}	$3s^23p\ ^2P^o_{3/2}$	(16.74±0.12) ms	LLNL EBIT	77
Si sequence				
Mn^{11+}	$3s^23p^2\ ^1D_2$	(11.16±0.10) ms	EKT	23
Fe^{12+}	$3s^23p^2\ ^1D_2$	(8.0±0.10) ms	HSR	78
Kr^{22+}	$3s^23p^2\ ^3P_2$	(6.82±0.1) ms	LLNL EBIT	79
P sequence				
Fe^{11+}	$3s^23p^3\ ^2P^o_{3/2}$	(1.70±0.02) ms	HSR	78
Fe^{11+}	$3s^23p^3\ ^2P^o_{1/2}$	(4.1±0.12) ms	HSR	78
Fe^{11+}	$3s^23p^3\ ^2D^o_{3/2}$	(18.0±0.1) ms	HSR	78
Fe^{11+}	$3s^23p^3\ ^2D^o_{5/2}$	(306±10) ms	HSR	78
Kr^{21+}	$3s^23p^3\ ^2D^o_{5/2}$	(0.80±0.03) ms	LLNL EBIT	80

(Table 2 concluded on facing page.)

Table 2. (*concluded.*)

Ion	Level	Lifetime τ	Trap type	Ref .
S sequence				
Ar^{2+}	$3s^2 3p^4\ ^1S_0$	(145±5) ms	HSR	81
Mn^{9+}	$3s^2 3p^4\ ^1D_2$	(18.02±0.16) ms	EKT	23
Fe^{10+}	$3s^2 3p^4\ ^1D_2$	(11.05±0.1) ms	HSR	78
K sequence				
Ca^+	$3p^6\ 3d\ ^2D_{3/2}$	(1.17±0.05) s	HSR	47
Ca^+	$3p^6\ 3d\ ^2D_{5/2}$	(1.09±0.05) s	HSR	47
		(1.100±0.018) s	RFT	82
Rb sequence				
Sr^+	$4p^6\ 4d\ ^2D_{3/2}$	(435±4) ms	HSR	46

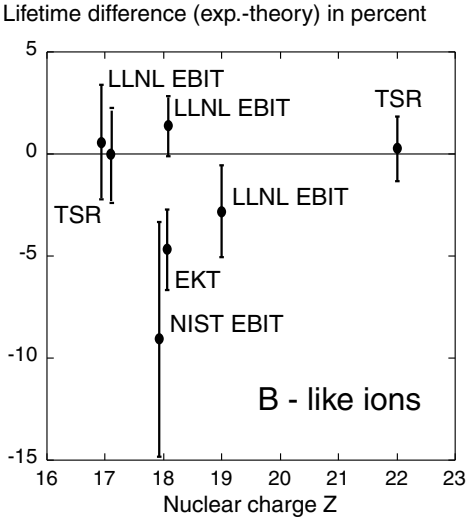
EBIT, electron-beam ion trap; EKT, electrostatic Kingdon-type ion trap; HSR, heavy-ion storage ring; RFT, radio-frequency ion trap.

raised many new questions, like the problem of how to explain a hot solar corona above a much cooler photosphere. Nowadays, forbidden transitions are a standard tool in plasma diagnostics [68] and astrophysics [69,70]. The strongest magnetic-dipole (M1) and electric-quadrupole (E2) transitions expected for many elements and their ions have been tabulated on the basis of experimental wavelength observations and semiempirical calculations [71]. 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 appropriate atomic lifetimes can be measured by a given technique. A selection of precise experimental results is given in Table 2, but a discussion of examples seems appropriate, too.

The simplest atomic system with a prominent M1 transition is the two-electron system with the transition $1s^2\ ^1S_0$–$1s2s\ ^3S_1$ already mentioned above. 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, few-ps lifetime), that is for lifetimes that span 15 orders of magnitude (see ref. 12). In between these extremes, ion traps, a heavy-ion storage ring, an electron-beam ion trap, a slow (recoil) ion beam, and foil-excited fast-ion beams have been employed, depending on the lifetime sought and the working range of the individual technique. The early 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 (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 generally does not surpass a few percent. Almost the only precise data available are those in the range $Z = 5$ to 7 obtained at the Heidelberg heavy-ion storage ring, TSR, [38,39] and those for $Z = 7$ through 10 obtained at the Livermore electron-beam ion trap EBIT-II [41–43], Fig. 2, Table 2. 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 then could decay either by the reverse process (autoionization, not showing any significant signal) or by radiative stabilization (DR). Then the ion would have changed charge state and could be detected (Fig. 1). The pioneering experiment on C^{4+} reached an uncertainty of 0.2% — a true benchmark. Data for N^{5+} and B^{3+} almost, but not quite, reached this precision.

At EBIT-II, electron energy modulation near excitation threshold was used in the pioneer experiment on Ne^{8+} [49]. In the later experiments on Ne^{8+} (and other ions), the electron-beam, after ion production, was switched off completely, converting EBIT to the operating conditions of a Penning trap [50]. The lifetime data for O^{6+} [41] and Ne^{8+} [42] turned out better than ±0.5%, ruling out all but two calculations

Fig. 3. Lifetime data for the $2s^2 2p\ ^2P^o_{3/2}$ level in the ground state of B-like ions from Cl^{12+} through Ti^{17+}. The data are normalized to the theoretical results given by Galavís et al. [89], which include a semi-empirical correction for experimental transition energies. A number of other theoretical results are within 0.6% of these values [71,90–93]. The experimental data other than for Cl XIII [52] are from an electrostatic Kingdon trap (EKT) [22], from the NIST and LLNL electron-beam ion traps [73, 87, 88], and from the heavy-ion storage-ring TSR (74).



that employ "exact" wave functions. For high nuclear charges, the predictions of these theoretical calculations, one nonrelativistic [83], one relativistic [44], diverge, as may be expected. However, the divergence is largely explained by the leading term of the relativistic correction, a factor $(1+1.07(\alpha Z)^2)$ [84]. Thus, the decisive point of calculational "quality" that distinguishes these calculations from many others is the detail of the wave functions. To distinguish between these good calculations in low-to-moderate charge state ions, an experimental precision of well under 0.1% would be required, that is, a level of precision that is not yet in sight. For nuclear charges in the medium-Z range up to $Z = 26$, existing experimental data of at best 5% uncertainty are inconclusive, although on average they agree slightly better with the relativistic prediction than with the nonrelativistic one. Data for $Z = 35$ and higher agree with the relativistic treatment; however, at uncertainties of typically a few percent, they only hint at the need for more than the leading-term relativistic correction to a nonrelativistic computation [85]. A single high-Z result with a very low quoted uncertainty [86] differs by more than the estimated experimental uncertainty from the prediction.

B- and F-like ions both have a $2s^2\ 2p^k\ ^2P^o$ ground term with two fine-structure levels (only the $J = 1/2,\ 3/2$ level sequence differs), giving rise to a single fine-structure transition that is prominent in solar spectra. Lifetime measurements have been tried using the combination of an ECR ion source and Kingdon trap at Texas A&M and at Reno [17, 21], at the NIST and Livermore electron-beam ion traps [52, 73, 87, 88], and at the Heidelberg heavy-ion storage ring [52, 53, 74]. The results of the Kingdon-trap experiments and of the experiment at the NIST EBIT agree with each other at the typically uncertainties claimed, but they clearly deviate from theory by much more than the error estimates. In contrast, the storage-ring results for Cl, Sc, and Ti (at uncertainties ranging from 3% to 1.5%), as well as the Livermore electron-beam ion-trap results for Cl, Ar, and K (again with uncertainties from 3% to 1.5%), are compatible with theory and with the expected iso-electronic trend (Fig. 3). Assuming — for good reason (as discussed above — storage of single-charge state ions under optimum conditions) — that the heavy-ion storage-ring technique yields data with the smallest systematic errors, there appear to be shortcomings in both the available Kingdon-trap and NIST EBIT data on these sequences.

The aforementioned $2s2p\ ^3P^o_2$ level in highly charged Be-like ions is predominantly depleted by an

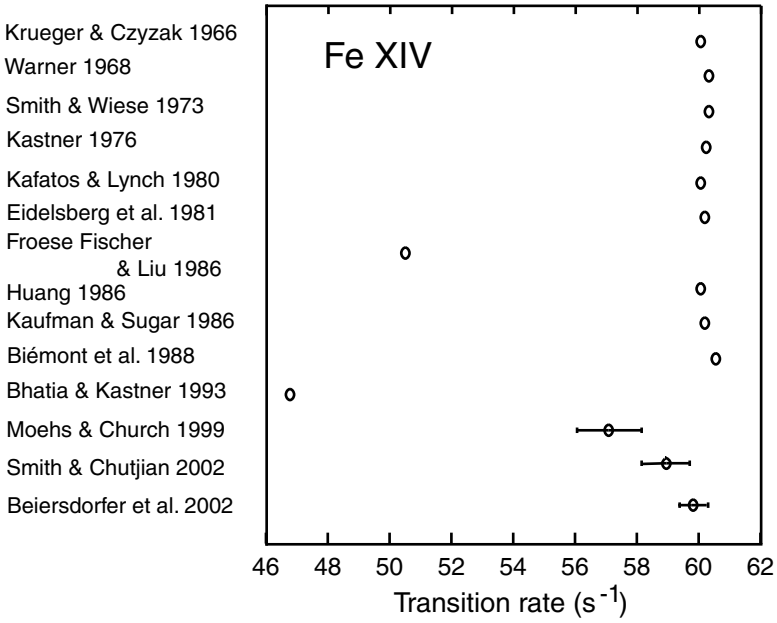
M1 decay to the 2s2p $^3P^o_1$ level. This case is interesting as it deals with an excited configuration for which calculations need much more effort than that for the ground configuration. The first example has been measured in Ar^{14+}, first at the electrostatic ion trap at Reno [22] and at the Oxford EBIT [72], and then again at the Livermore EBIT-II device [73]. At a nominal precision of 5%, the Reno result is not in agreement with most, including very recent theory [94]. As the latest calculation produces almost the correct fine-structure intervals from an ab initio calculation, before it is then semiempirically adjusted, it would be surprising if the clearly different experimental finding for the lifetime would be validated by another measurement. The two experiments on this level lifetime that have employed electron-beam ion traps yielded lifetime results that at face value are not more precise than those obtained with the Kingdon ion trap at Reno. However, they agree with each other and they overlap, within their error bars, with theory. While both types of experiment suffer from a relatively low signal rate (the excited electron configuration evidently is less efficiently excited than the levels of the ground complex), the electron-beam ion-trap experiments may well have better control of systematic errors, including the important ion-storage-time correction. What is needed is a storage-ring measurement on this system, because there the control of such parameters would be best. Time will tell which of the present data will last.

C- and O-like ions are another 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 with 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). For low-charge-state ions the E2-decay branch of the $^1S_0$ level to the $^1D_2$ level dominates, while for higher charge states the M1 decay to the $^3P_1$ level outweighs the others by orders of magnitude. A recent storage-ring measurement [76] fixes the lifetime in F^+ (near 425 ms) to $\pm 3\%$. Corresponding measurements on C-like O^{2+} and F^{3+} have very recently been done as well [59]. For the lower lying $^1D_2$ level, the wavelength "window of opportunity" 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 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% [75]. This is clearly less than the scatter of various predictions or the 10% uncertainty quoted for more recent, quite extensive calculations.

The ions of the N sequence are more complex than the above ones, with a $^4S^o_{3/2}$ ground term in the $2s^22p^3$ 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-milliseond ($^2P^o_{1/2,3/2}$), many-millisecond ($^2D^o_{3/2}$), and few-second ($^2D^o_{5/2}$) ranges, respectively. Experiments at the TSR heavy-ion storage ring (UV range observations only) have corroborated theory for the first three of these level lifetimes in S^{9+} [59]. The interesting multisecond 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.

In solar corona and other astrophysical plasma spectra, forbidden lines of Fe in medium-charge states figure prominently. These are ions with an open 3p shell. In the ground complexes of these $n = 3$ shell ions, the same level structure as in B-like to F-like ions is repeated. An additional challenge for theory lies in the need to deal with another closed shell. This combination of interests has incited a series of measurements by the Texas A&M group setting up an experiment at Reno (using an ECR ion source and a Kingdon trap [17, 22–24], and concentrating their measurements on Ar ($Z = 18$), Mn ($Z = 25$), and Fe ($Z = 26$). In one of their reports, the authors state that the comparison of their experimental results (some claimed with errors as small as 0.9%, some with 15% uncertainty) with theory is inconclusive. Considering that the calculations carry much larger uncertainty estimates (if any are given at all, typical estimates run to 20–50%), this might have appeared as good news for the astrophysics community, replacing the vagaries of theoretical results with firm experimental data.

Fig. 4. Transition-rate data for the $3s^2 3p\ ^2P^o_{3/2}$ level in the ground state of the Al-like ion Fe^{13+}. Note how most calculational results agree within a close interval with each other. The two deviating theoretical results apparently have not been adjusted for the experimental fine-structure intervals, clearly underlining the computational problems with that entity. The first (Texas A&M/Reno) electrostatic ion-trap measurement (by Moehs and Church) [24], however, did not resolve the evident discrepancy. The electron-beam ion-trap measurement at Livermore (by Träbert et al.) [77] clearly agrees with the (semi-empirically adjusted) majority of the predictions and settles the case, while the second (Caltech) electrostatic ion-trap measurement (by Smith and Chutjian) [95] has a result in between.



However, within very few years after the Reno experiments (on an electrostatic ion trap), experiments employing the heavy-ion storage ring at Heidelberg and the electron-beam ion trap at Livermore have revisited a number of the Fe levels covered in the Reno work (and extended the coverage to other ions and levels) [77, 78] (Fig. 4). The newer data seem to reflect better control over experimental conditions, resulting in raw data that leave little uncertainty in the way of interpretation and evaluation. Consequently, most of the newer results carry much smaller uncertainties. Very notably, however, they often differ significantly (by multiples of the stated errors) from the Reno results. Moreover, in some cases consistency checks against the predicted pattern of lifetimes of a group of levels was possible, and the new experimental data matched the expected pattern. For lack of consistent theoretical coverage, no sweeping statement as to "agreement with theory" was possible, but calculations could be identified that for good reason appeared to be more reliable than others, and the results of which were compatible with experiment. As a consequence, the aforementioned "inconclusive" comparison of the Texas A&M Reno results with theory (as presented by the Texas A&M Reno group) appears to be likely the fault of the insufficient reliability of the Reno experiments. Some of the Reno results probably are very good, but how to identify the reliable ones among the apparently overstated ones?

The low particle density in electron-beam ion traps permits the observation of light from levels with millisecond lifetimes, like forbidden optical transitions in many highly charged ions. Cycling the electron-beam energy between "on" for ion production and "off" for observations of the trapped ions in the magnetic trapping mode [50], transition rates for various M1 and M2 transitions have been measured, for example, for various ions of Kr [80]. The M2 transitions in this case (Kr^{18+}) were not observed

directly (for reason of spectroscopic access), but their (almost 50%) influence was deduced from the decay curves of other M1 decays of the same upper levels. Only after accounting for the M2-decay branches, the calculated level lifetimes agreed with the measured ones.

Electron-beam ion traps are 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 [96]. However, even in the ultrahigh vacuum system of an electron-beam ion trap there are contaminations, and there are many ionic systems for which no theoretical data are available to help proceed with line identification. This field of forbidden lines in the spectra of highly charged ions will require years of further work that will greatly benefit from an electron-beam ion trap's unique feature of "dial-up a (maximum) charge state".

In general, fine-structure intervals increase rapidly with ionic charge. A rare exception has been found in the Ti iso-electronic sequence. The $3d^4\ ^5D$ $J = 2$–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 by possibly using many different elements with little change to the interferometer optics. After initial work at the NIST EBIT, the world-wide search for this transition has meanwhile reached up to Bi^{61+} [97–100]), and the transition rate has been determined in Xe^{32+} [101] at the NIST EBIT. The result, a lifetime value of 2.15 ms with a 7% uncertainty, comes close to that of a (semi-empirically scaled) calculation.

Among the low-charge many-electron 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 has been measured by a number of groups, but not always in agreement with each other, to a precision of a few percent. At long last, a heavy-ion storage-ring measurement, using CRYRING at Stockholm, has been added to the pool of ion-trap data. While the authors claim that their observations resolve earlier inconsistencies among experimental data [47] (some of which carry a smaller error bar), this storage-ring measurement has not yet reached a precision of better than 5%. This is because of the long (1 s) atomic lifetime and of the problem of survival of metastable ions in an environment that features an ion storage time of the injected 40 keV beam (dominated by ground-state ions) of about 45 s. Notably, the new lifetime results for the two fine-structure levels differ much more from each other (8%) than expected from theory (typically 2%, see the literature survey in ref. 102), and this indicates the need of further work. Since then, the Mainz group (now using a linear Paul trap) [82] has confirmed the presence of an earlier systematic error in their experiments that slightly shifted the $J = 5/2$ level lifetime. Their new result for this level is in perfect agreement with the storage-ring result, but bears a smaller error bar of only 2%. This shifts the interest back to the $J = 3/2$ level that came out so differently in the storage-ring work.

The CRYRING group specializes in lifetime measurements of singly charged heavy ions [46, 47]. Some of these ions are of particular astrophysical interest (Fe^+ [103]), others are challenging because of very long atomic lifetimes (about 5 s in La^+ [104]). There is one interesting practical difference in the data-taking of this laser work at CRYRING Stockholm and the aforementioned "passive" detection used at the Heidelberg storage ring: The laser light changes the population of the level of interest, so that the remaining level population can sensibly be probed only for one given (adjustable) time into the injection cycle. Decay curves are thus being built at one point per cycle, depending on the constancy and reproducibility of the ion production and injection process. With the passive observation of the radiative decay used at Heidelberg, the detection efficiency is low (for geometrical reasons), but data are being filled into the full decay-curve data set in each cycle, averaging out any source variations from the beginning. The active influence the level populations by laser light, however, has a selectivity that can resolve hyperfine structure levels in singly charged heavy ions [46, 48].

A classical case for studying the influence of the hyperfine structure on atomic lifetimes is the $1s2p\ ^3P^o_0$ level in He-like ions that normally cannot decay to the $1s^2\ ^1S_0$ ground state, because no single-photon $J = 0$–0 transitions are allowed. However, as was recognized over 60 years ago [105],

with a nonvanishing nuclear spin, there may be hyperfine components that have the same total angular momentum quantum number $F = J$ (electron shell) $+ I$ (nucleus) as some neighbours. 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). Beam-foil spectroscopy has used this effect in various He-like ions. In ions with ground-state transitions that do not involve 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 [106, 107] — too small for beam-foil work, but probably just right for ion traps — all across the periodic table.

There is at least one heavy ion, In^+, that features a hyperfine-induced decay in the working range of the existing detection equipment at the Heidelberg storage ring. 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 [108]), but it also has a 5s5p $^3P_0^o$ level with a hyperfine-structure-induced decay at a wavelength near 236 nm and a lifetime of (195 ± 8) ms [108]. Theory is within about 30% of the experimental data in both cases [109]. A precise lifetime measurement calls for a heavy-ion storage ring in this case, and the passive observation technique practiced at Heidelberg would seem applicable. However, the Heidelberg heavy-ion storage ring has no ion-beam diagnostics for sub-MeV heavy-ion beams yet.

The opposite problem, a heavy ion with only a single electron, has already been tackled, including lifetime measurements, with $^{209}Bi^{82+}$ [110] and $^{207}Pb^{81+}$ [111]. In these hydrogen-like ions, the hyperfine-structure transition in the ground state (corresponding to the 21 cm line of hydrogen used for masers and radio astronomy), because of the Z^3 scaling of the hyperfine-structure splitting, is a transition in the ultraviolet region of the spectrum that can be pumped by visible laser light, using the massive Doppler shift that is available with high-energy ion beams stored in the ESR storage ring at GSI Darmstadt. The first lifetime results on these ions were not yet precise in the sense of some of the measurements discussed above, but in the second round, the measurement on Bi yielded a result with only 0.4% uncertainty [112] and in agreement with a calculation that includes the nuclear-magnetic-moment distribution in the nucleus — a measure different from the wavelength measurement that also helps to understand this nuclear property.

5. Conclusion and prospects

The new storage-ring results on intercombination and forbidden transition rates in few-electron ions are more precise than present calculations, even after semi-empirical corrections of the latter, and they thus provide a long-missed challenge to theory. Obtaining any such data in the first place was a challenge for experiment, which has first been met by conventional ion traps. These traps have served physics well, but they have many intrinsic problems (most notably the vacuum working range) that preclude the gathering of truly precise lifetime data. Heavy-ion storage rings seem best suited for this task, as they offer separate optimization of ion production, charge-state selection, ion storage, as well as the best control over ion-loss rates. Lifetime error margins of better than 0.2% have been reached by several experimental techniques at the TSR heavy-ion storage ring, and further improvements of this precision appear quite feasible. Development work, however, is needed to increase the supply of highly charged ions to the present storage rings. Small storage rings, like ELISA at Aarhus, seem ideal to extend the versatility and precision to the lifetime study of light molecules. Electron-beam ion traps like EBIT-II at Livermore do well for precise lifetime measurements on few-electron ions in the X-ray range. However, problems have been encountered when aiming at high-precision lifetime data in the visible range [113]. Nevertheless, studies to identify and quantify or remove systematic errors have meanwhile led to the first measurements at the 1% level of uncertainty also for lifetime measurements on optical transitions [77]. Provided systematic errors are under control, as seems to be the case at the Livermore and Oxford devices, an electron-beam ion trap has the enormous advantage of versatility,

and of being applicable to many elements, in particular to high-charge states.

However, there are always some trade-offs: In the storage ring, ions of only a single-charge state are present, and little spectral discrimination is needed, because in many cases there will only be one (or at worst a few) long-lived levels that decay by radiation to which a given detector is sensitive. This advantage balances the drawback that only a fraction of order 10^{-3} of the storage-ring circumference is seen by the detection system. (This is not a problem for the active probe technique used at CRYRING, but then their experiments depend on lasers that simply are not available for probing highly charged ions.) Also, Doppler broadening is not a problem, if there are no nearby foreign lines. With an electron-beam ion trap, in contrast, the complete light source can be imaged onto a detector, and because of the smallness of the light source, it is possible to do high-resolution spectroscopy without any (narrow) spectrometer entrance slit. However, this spectral resolution is also necessary, because there is a distribution of charge states in the trap, and there are cooling-gas admixtures and impurities. It is then that the limited efficiency of the spectrometer reduces the overall efficiency. With either light source, it takes about two days to record decay curves of meaningful statistical value. This data rate is better than that from many conventional ion-trap experiments by more than an order of magnitude. Moreover, the combination of smaller systematic errors with much better signal statistics results in a notable improvement in overall precision. Meaningful precision in this context is better than 1% in the X-ray range and aiming for 1% (or better) in other spectral ranges with their notable detector noise, all for lifetimes between roughly 0.1 ms and a few hundred ms. Similar levels of precision may be expected from new developments on the laser interrogation of ions stored in classical ion traps [82, 114]. Five years ago these small error margins seemed only distant goals, but now some of this is a near-routine operation, keeping in mind the next factor of 3 to 5 in better precision.

Producing reliable and precise data as presented by many of the data listed in the tables in this review has meant an important step forward in the understanding and theoretical description of atomic structure and dynamics. The further insight to be gained now, from even better atomic-lifetime measurements using ion traps of all sizes, is not yet estimable. In this field, there is a challenge of improvement to both theory and experiment. For quite some time, theoretical initiatives for more extensive calculations have been stymied by the criticism that "there is nothing to compare better calculations with". This claim has now become obsolete by the leap in experimental precision. Theoreticians have not been idle, though. There are 20-year-old results that coincide with the new measurements — but at the time, the calculations were expected to be not more reliable than, say, 20 to 50%. This uncertainty estimate has nowadays shrunk, thanks to a variety of factors, among which are cheap computing power that permits vast wave-function expansions, systematic checks for convergence, and some new ideas as well [115], say, 1% to 10% for intercombination and forbidden transition rates. As mentioned above, the science case is open: the most precise lifetime measurement [45] and the most precise calculation [55], both done for the intercombination transition in C^{2+}, disagree by more than their combined error estimates.

For some forbidden transitions in few-electron ions, the majority of calculations has provided very reproducible results that are now corroborated by experiment. Here, the calculations seem to have been more reliable than the intrinsic error estimates (of 10% or more, if given at all) had led us to believe (Fig. 3), this has not prevented some recent calculations from straying off the established — and now corroborated — range. It has to be kept in mind, however, that fine-structure intervals in multi-electron heavy ions still defy ab initio calculation at the 5 to 10% level, a problem that on its own carries an uncertainty for M1-transition rates that is three times as large. A warning example is shown in Fig. 4. Therefore, many calculations use, at some stage or other, semi-empirical adjustments to experimental term values. The smaller uncertainties quoted above then often relate to such adjusted calculations, but rarely to ab initio results. While the foundations of quantum mechanics are undisputed, their implementation into computer codes to achieve precise and reliable quantitative predictions on many-body problems, like ions with several electrons in the valence shell, still has a long way to go. The precise lifetime data now obtainable at heavy-ion storage rings and at the Livermore electron-beam

ion trap provide important benchmarks for this quest.

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