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Cascade problems in some atomic lifetime measurements at a heavy-ion storage ring

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Abstract. Lifetimes of $3s^2 3p^k$ ground configuration levels of Al-, Si-, P-, and S-like ions of Fe, Co, and Ni have been measured at a heavy-ion storage ring. Some of the observed decay curves show strong evidence of cascade repopulation from specific $3d$ levels that feature lifetimes in the same multi-millisecond range as the levels of the ground configuration.

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1. Introduction

After many decades of atomic spectral analysis with the goal of understanding atomic structure in ever more complex atoms and highly charged ions, a notable part of the spectroscopic effort has moved to the application of spectroscopic knowledge to the understanding of the light emitting plasmas, be they terrestrial or astrophysical. Collisional-radiative modeling involving thousands of levels and tens of thousands of transitions is employed to synthesize spectra that can be compared to actual observations, trying to extract information on plasma density and temperature from specific line intensity ratios. By necessity, most of the information used in the models, especially for transition rates, is theoretical. For a number of prominent electric dipole transitions, beam-foil spectroscopy [1] has tested the predictions of such rates to within 5 to 15%, with only a few lifetime measurements being more accurate than 3%. One of the serious limitations of accuracy in beam-foil lifetime measurements is the occurrence of significant repopulation of levels of interest by cascades from higher-lying levels that are easily populated in the ion-foil interaction. The initial population takes place at high

density (solid state density inside the exciter foil) and is then modified by low-density population effects - including capture into high-lying levels - near the rear surface of the foil. If the time structure of the cascades that feed a given level can also be measured, a correlated analysis (evaluation of Arbitrarily Normalized Direct Cascades - ANDC) [2] can yield the the primary (cascade-fed) level lifetime with high accuracy, sometimes with a precision of about 1%. However, in most cases, the available spectroscopic equipment cannot reach all of the important cascades, or the lifetimes involved are shorter than the time resolution of the detection system. Then cascade modeling may help to a degree; such modeling usually is based on parametrizations of level populations, lifetimes and branching fractions. In a number of cases it turns out that relatively long-lived levels in low-lying configurations (with level lifetimes, say, in the range of hundreds of picoseconds to many nanoseconds) can be measured best, because most cascades have lifetimes of a few picoseconds and will boost the intensities of the slow decays, but do not much distort the decay curves. However, it has been recognized long ago that in quite many atomic systems there are peculiar higher-lying configurations with individual levels that break the usual lifetime pattern in that they feature very long lifetimes, too [3]. Such slow cascades might distort the tails of long-lived levels that previously had been assumed to be unaffected [4].

Decades later, the techniques of beam-foil spectroscopic lifetime measurements were extended by employing heavy-ion storage rings (see, for example, [5, 6, 7, 8, 9, 10, 11, 12] or reviews [13, 14, 15]), addressing lifetimes of levels in low configurations that would decay only by spin-forbidden decays, or ground configuration levels with electric-dipole forbidden decays. The new range of lifetimes, some 5 to 10 orders of magnitude longer than treated before, gave rise to the optimism that all high-lying levels were much shorter lived than the low-lying levels of principal interest, so that slow cascades would surely not matter, and that consequently this systematic error that plagues beam-foil spectroscopy would not spoil the remarkably accurate results obtained in a number of measurements made at heavy-ion storage rings. In fact, electron beam ion traps [16, 17] offer a second route to similar levels with millisecond to second lifetimes, and careful experiments have helped to sort out various error sources. With sufficient care being taken, the results were found to mutually agree among the two techniques [18]. Two lifetime results since obtained at the Heidelberg electron beam ion trap [19, 20] have claimed unprecedented accuracy, which, if valid, would even test a QED correction to the transition operator that amounts to 0.45%. At the claimed high accuracy, however, these experiments disagree with results from a combination of highly developed quantum mechanics calculations and state-of-the-art QED, and this discrepancy renews the urge to understand all significant sources of systematic error in such measurements.

Apart from these two Heidelberg EBIT experiments [19, 20] with their 0.1% lifetime error estimates, most of the better lifetime measurements using a heavy-ion storage ring or an electron beam ion trap have uncertainties near 1% (see the aforementioned reviews). The bulk of these measurements have shown that various results from conventional ion traps must have suffered from unrecognized systematic error. In a

single case, a result from the heavy-ion storage ring, on Fe XIV [21], did not agree with results from electron beam ion traps [20, 22]; the unexpected 10% deviation was noted as problematic and was tentatively blamed on a poor signal-to-noise ratio, because the ion beam current in the heavy-ion storage ring had been much lower than with the other measurements of Fe ions [21]). A recent report on lifetime measurements using an electrostatic ion trap [23] referred to this particular, somewhat suspicious, Fe XIV lifetime result from the heavy-ion storage ring as if it did not carry a warning label [21]; maybe in comparison the result from the electrostatic ion trap looked less poor.

Instead of redoing the heavy-ion storage ring experiment on Fe XIV, we have elected to extend the measurement along the isoelectronic sequence, in order to study systematic errors while avoiding a remeasurement that might possible suffer from an accidental line blend. As it turns out, the results on Al-like ions of Co ($Z = 27$) and Ni ($Z = 28$) have a much better signal-to-noise ratio than the earlier measurements of Fe, but they show the same systematic deviation from reliable predictions. Similarly, we have extended lifetime measurements on Si- and S-like ions of Fe to these two elements, and although working under somewhat different experimental conditions compared to the earlier observations of Fe, we have again found additional decay components in the same range of millisecond lifetimes as the levels of primary interest, and these cascades must arise from $3d$ or even higher levels. Apparently the problem of slow cascades, which has plagued beam-foil spectroscopy for decades, has resurfaced, even at this very different time scale.

In this report, we present the evidence and identify, corroborated by our own calculations, the likely levels of origin of the cascades. For our $3s^2 3p^k$ ground configuration levels of interest, most of these slow cascades originate from specific high-J $3d$ levels. However, in ions of many isoelectronic sequences, one can find even higher lying levels that may give rise to slow cascades or act as population traps in actual plasmas (see the example of Ni-like ions [24, 25]). Metastable levels play an important role in many collision experiments; some of the levels we discuss may contaminate such experiments at heavy-ion storage rings, because their long lifetimes might let them survive even periods of electron beam cooling. The enormous computational effort in treating those levels usually has precluded their inclusion in radiative-collisional models so far. Our observations indicate that this neglect may be rather detrimental to the modeling value.

2. Experiment

Our experiments were performed at the TSR heavy-ion storage ring at MPI-K Heidelberg, using the same techniques as described before [6, 7, 8, 11, 12, 21, 26]. A 10 MV tandem accelerator equipped with a sputter ion source provided an ion beam of defined elemental species, charge state and energy. This ion beam was guided by magnetic fields to the 55-m circumference storage ring where the beam current was increased by stacking to about 30 times the current of the injected beam. Then the injection was stopped and the beam left coasting. Passive observation of photons

through a window in the ultra-high vacuum vessel yielded a decay curve as a function of storage time. After 200 ms, when in most cases the signal level had disappeared in the detector background, the measurement was stopped, the ion beam was dumped and the storage ring filled again. The data were sorted in 1 ms intervals and stored every minute, permitting to reject afterwards and offline the data records for those times when the ion beam had failed. Within a measurement day, the accumulation time per data channel amounted to about 5 min. As detectors, a solar blind photomultiplier (EMR 541Q, 'Q tube') with a dark rate of about 1 cps and a photomultiplier for near-uv and visible light (EMR 541N, 'N tube', dark rate about 10 to 15 cps) were available. The latter detector was used with a narrowband interference filter in the light path. Although vacuum gauges in the storage ring sector of the experiment were shut off and windows covered, some daylight must have leaked through windows in far-away sections of the ring vessel. This was clearly seen in the background rate changing with the day and night cycle and the cloud cover. Nighttime measurements had a much superior signal-to-noise ratio. With the daylight spectral maximum near 550 nm and the N tube spectral sensitivity maximum near 350 nm, spectral lines in the blue were also observed with a better signal-to-noise ratio than those in the green, yellow or red, which along an isoelectronic sequence relates to better or worse conditions for the observation of specific elements.

When recording optical decay curves of an ion sample, one has to monitor the number of ions in the trap, or the ion loss rate. In the present experiments, the ion storage time (confinement time constant) was measured beforehand; at storage time constants of typically 10 to 20 s, the corresponding correction to the observed decay rate was of the order of 0.1% in most cases studied here, which practically balanced the correction for relativistic time dilation. Moreover, the ionization of the residual gas in the storage ring vessel, effected by the coasting ion beam, was detected by a beam profile monitor and stored alongside the optical signal. Of course, the diagnostics equipment at such a heavy-ion storage ring is designed to match the usual operational mode of storing ions for long times; there is no truly reproducible probe for the ions in the first, say 20, milliseconds of a storage cycle. The optical signal indicated that the stored ion beam was well behaving after about 1 ms from the time of injection, much more reliably than the diagnostics equipment.

There was one procedural difference to the Fe measurements of a few years ago [21, 26]. In those measurements, the ions reached charge states q=8+ to 12+ in a single (foil) stripping process in the high voltage terminal of the tandem accelerator; this stripping thus took place at an ion beam energy of about 10 MeV, and the ions were then accelerated to energies of about 70 to 140 MeV. The charge state distribution did not extend to make sufficient quantities of Fe ions with q=13+ this way. For this ion species, a gas stripper was used and ions of a lower charge state ion were accelerated, which were then stripped (at an energy of about 28 MeV) by passage through a foil after the accelerator. This latter procedure was employed for all the (higher) charge states that had to be reached in Co and Ni with their higher nuclear charges; however, the foil

stripping energies were closer to 40 to 60 MeV in these cases.

3. Calculations

The interpretation of the experimental data has been helped by various calculations that in general have been described elsewhere. Among the program packages used is the Fast Atomic Code (FAC) developed (and used for some contributions to the present project) by M F Gu [27] which also permits radiative-collisional modeling (P Beiersdorfer providing helpful numerical examples); both calculations concerned Al-like ions. The group of Y Ishikawa has developed a Multi-Reference Møller-Plesset code to treat the structure of many electron ions with high accuracy [28, 29, 30, 31]. Earlier applications of the code have targeted, for example, individual states of Si-like ions. This work has now been expanded to cover all levels of the ground configuration and a few selected long-lived states of higher configurations, as well as similar states in S-like ions. This work will be presented in more detail and with many results elsewhere [32], so that we elect to show only a few specific results in the present study, along with the experimental findings.

4. Experimental data and interpretation

In preceding experiments on iron group elements at the heavy-ion storage ring, iron ions Fe^{6+}, Fe^{8+}, Fe^{9+}, Fe^{10+}, Fe^{11+}, Fe^{12+}, and Fe^{13+} have been studied [21, 26]. In particular for the isoelectronic sequences of P and Cl, the investigations have been expanded to other ions of iron group elements (up to Cu) and to some 3d levels [12, 33, 34, 35], and very recent work has also addressed hyperfine-induced and M2 decay channels in Mg-like ions of Co, Ni, and Cu [36]. In the context of slow cascades, the isoelectronic sequences of Al, Si, and S appear to be most significantly affected, and we address our measurements on ions of these sequences in the following. The experimental parameters are listed in table 1.

4.1. Al-like ions

By basic atomic physics principles (and in the single-configuration, non-relativistic limit), the dominant M1 transition rate (more than 99.9%, with only a weak admixture of E2) of the fine structure transition in the ground term $3s^2 3p\ ^2\mathrm{P}^\mathrm{o}$ is expected to be given by the line strength S=4/3 and the third power of the transition energy. The transition energy has often been calculated but poorly, and with very few exceptions (e.g., [28]), the experimental value of the transition energy (from a spectroscopic wavelength measurement) has been used instead (the process usually is called a "semiempirical adjustment"). The measured wavelength encompasses the QED correction of the level energies that otherwise would be called for; this correction has an effect on the level lifetime that is of the order of 0.4% [19]. However, there is also a QED effect that applies to the M1 transition operator (see discussion in [19]) and that has a 0.45%

effect on the level lifetime. It is not yet clear how accurately such level lifetimes can be measured (the Heidelberg EBIT group claims an overall uncertainty of about 0.1% [20] for their (16.726 ms) measurement on Fe XIV, whereas the preceding Livermore EBIT measurement [22] (16.74±0.12 ms) reached an uncertainty of 0.6%). A measurement that deviates by more than 1% from the simple prediction, however, is certainly suspect. Such measurements (all for Fe XIV) are the two Kingdon trap (electrostatic ion trap) measurements performed at Reno [37] (17.52±0.29 ms) and at Caltech [23] (17.0±0.2 ms), and our own earlier TSR experiment [21] (18.0±1.2 ms).

The observations of the M1 transitions in Al-like ions of Co and Ni employed the aforementioned N tube and interference filters of band pass 431-435 nm (high transmission T=70%) for Co^{14+} and mean wavelength 360 nm, bandpass interval 10 nm (transmission not documented) for Ni^{15+}, respectively. Our decay curves obtained with Al-like ions of Fe (previous work), Co and Ni (new work) appear to be well represented by a single exponential (figure 1); however, the fits return a lifetime value that deviates from the above prescription by about 9%. After the first measurement, it seemed possible (though rather unlikely) that another long-lived level in the same ion species (the ion beam guidance system of the storage ring is highly selective) might cause a spectral blend, or that the result was a statistical fluke in the face of a not very good signal-to-background ratio. Finding the same deviation for two more ion species (at different transition wavelengths and therefore with different interference filters, and with much better signal statistics), we see the only likely cause in a cascade that is somewhat slower than the principal decay, but still rather close in lifetime.

A possible candidate, the long-lived $3s3p3d\ ^4F^o_{9/2}$ level, has already been discussed in the report on the earlier measurements on Fe XIV [21], when no theoretical lifetime value was available yet. By the time of the Livermore EBIT experiment [22], a level lifetime of 19 ms was calculated using the FAC code. Recent FAC calculations by Gu [38] point at a level lifetime of 32 ms; however, none of these calculations has been undertaken with the attention to detail (complexity of wave function) that may be required for a definitive calculation. Our own MB-MP calculation also returns a 19 ms lifetime. If the principal ($3s^23p\ ^2P^o_{3/2}$) and cascade ($3s3p3d\ ^4F^o_{9/2}$) level lifetimes differ by as little as the 16.6 ms established for the former and the 19 ms estimated for the latter, no multiexponential fitting process will be able to disentangle the two contributions. If the cascade lifetime amounts to the 32 ms of the more recent calculation, the chance may be slightly better, although a rule of thumb requires that the two components be different by at least a factor of three. We note in table 2 that the calculations indicate slightly different isoelectronic trends for the two lifetimes (see figure 2), and that in more highly charged ions, a resolution of the decay curve blend by multi-exponential fits may be easier to achieve.

In the low-density environment of an electron beam ion trap, the $3s3p3d\ ^4F^o_{9/2}$ level is not populated very much. FAC calculations at the time [22] have indicated a very small excitation cross section, but that is only part of the story. Any of the more easily excited higher-lying levels may decay towards lower-lying levels, possibly including the

$^4F^o_{9/2}$ level, and FAC indicates a level population that may be as high as 0.5% of that of the ground state (at low density) ([39]). Under the high-density conditions inside a stripper foil, the population of the levels of a swift ion is distributed over many levels; when the ions exit the foil, the rapid excitation ceases, and the populations rearrange, with long-lived levels (and the ground state) acting as population traps.

Without time-dependent collisional-radiative models in reach, we cannot take guidance from modeling, but we do so from experiment. In a first step, we simulate decay curves based on the calculated primary lifetime ($^2P^o_{3/2}$) and a second component that represents the $^4F^o_{9/2}$ level, with different relative amplitudes, plus a realistic background. We then fit a single exponential to this composite curve. For an apparent lifetime close to that seen in the experiment, we find that the cascade amplitude (with a cascade lifetime from FAC) is about 9% of that of the primary, reflecting about the same as a population ratio after foil excitation. We note that the cascade level decays via several distinct cascade paths; our simple model captures only the fraction of cascades that reaches the $^2P^o_{3/2}$ level. We then invert the process and try out whether with such relative amplitudes and calculated lifetimes as start parameters, a fit of two exponentials finds any stable solution. For a few of the data sets this is, indeed, the case, suggesting that the actual $^4F^o_{9/2}$ level lifetime is about halfway between the recent FAC predictions and the lifetime of the $^2P^o_{3/2}$ level. However, the fits show considerable ambiguity (large uncertainty estimates) because of the proximity of the two time constants involved. Therefore the multiexponential fits demonstrate consistency, but such data are insufficient as quantitative tools, and we cannot ascribe realistic error estimates to most of such fit results (see table 2). However, a multiexponential analysis of our Ni data (where primary and cascade lifetimes are expected to differ by almost a factor of two) comes close to the MB-MR calculated lifetimes of primary and dominant cascade levels. A future measurement on Al-like Cu or Zn might provide even clearer evidence on the two major decay components and thus on the cascade situation after ion-foil interaction.

We can discuss in such detail only our own measurements and evaluations, but the observations have implications for the accuracy of other experiments, too. Whenever - in such relatively simple cases - the measured lifetimes exceed the prediction, cascade repopulation should be considered. If the measured lifetime is too short, previous experience [18] has pointed to insufficient control over ion loss from the sample under observation. Unfortunately, some groups working with conventional ion traps have reported results on either side of the likely correct values, which suggests further, unrecognized or underestimated systematic errors in that work. If, in the context of the present study, we limit the discussion to the measurements on the Al-like ions, we see that the storage ring result [21] differs most (by some 9%) from the expected lifetime result. The data obtained using an electron cyclotron resonance ion source (ECRIS) and an electrostatic ion trap [23, 37] differ from expectation by a few percent, all three by more than their perceived 1-σ uncertainty. The Livermore electron beam ion trap result overlaps with expectation within the 0.6% error, while the Heidelberg EBIT result with its 0.1% stated error is almost 1% away from expectation. All of these

results show a lifetime that is longer than expected. The highest density environment (foil excitation) seems to result in the largest deviation and the low density environment in an EBIT in the smallest. The storage ring decay curve simulation including the single recognized long-lived cascade level suggests a level population that amounts to about 9% of that of the $3s^2 3p\ ^2\mathrm{P}^\mathrm{o}_{3/2}$ level. The electron beam ion trap offers the lowest density environment of the three types of experiment. Estimates based on a radiative-collisional model [39] suggest a cascade level population of the order of 0.2% to 0.4%. (In [22] only the excitation cross section to the specific cascade level was considered, but the level population is a result of exciting and reshuffling many levels, which is why the relative effect on a particular level can be much higher.) Our simulations then indicate an effect of similar size (0.2 to 0.4%) on the apparent level lifetime of the $3s^2 3p\ ^2\mathrm{P}^\mathrm{o}_{3/2}$ level. For the Livermore measurement, such a correction would remain inside the stated error bar, while the Heidelberg result would possibly be affected by a multiple of their stated uncertainty, reducing the discrepancy with expectation. For the Reno and Caltech work (ECRIS and Kindon-type ion trap), the offset of their results is suggestive of a $3s3p3d\ ^4\mathrm{F}^\mathrm{o}_{9/2}$ level population that amounts to several percent of that of the $3s^2 3p\ ^2\mathrm{P}^\mathrm{o}_{3/2}$ level. It will be difficult to actually measure these small percentages in independent experiments.

Figure 2 demonstrates that the cascade problem is most difficult to deal with for Al-like ions of elements near $Z = 26$ (Fe). For all practical purposes, the M1 transition rate in the ground term of Al-like ions should therefore be taken from the above "semiempirical" recipe. This should not preclude, of course, further study of systematic error sources with heavy-ion storage ring and electron beam ion traps which in many other cases have delivered accurate lifetime results. It would be interesting to study an ion species that can be produced by either foil or gas stripping in the injector accelerator serving a heavy-ion storage ring, and to find out whether there is a visible cascade influence and how it differs between foil and gas excitation.

4.2. Si-like ions

In the $3s^2 3p^2$ ground configuration of Si-like ions, various transitions are of astrophysical interest, but wavelengths and lifetimes constrain any measurements of level lifetimes to a few elements each. We have extended the measurement of the $3s^2 3p^2\ ^1\mathrm{D}_2$ level lifetime from Fe [21] to Co and Ni, watching out for the cascade contributions from the almost equally long-lived $3s^2 3p3d\ ^3\mathrm{F}^\mathrm{o}_4$ level (see discussions in [26, 29]. None of the numerous decay branches of that level have ever been observed, but the cascade feeding - the strongest decay branch points to the $3s^2 3p^2\ ^1\mathrm{D}_2$ level - is evident in the decay curve of the latter level. In Fe^{12+}, the lifetimes of principal decay ($^1\mathrm{D}_2$ level) and cascade are almost identical, and individual values for the two decay time constants have been obtained by modeling in combination with multiexponential fits [26]; the error estimates based on such a procedure may be optimistic. Fortunately, the isoelectronic trends of the two level lifetimes differ [29], so that the situation is easier to disentangle in elements heavier than Fe, and direct multiexponential fitting becomes viable. The

new decay curve data are consistent with the picture gained before and corroborate the calculations.

In Co^{13+}, both decay branches of the $3s^2 3p^2\ ^1D_2$ level were observed. The 311 nm line (all wavelengths taken from the online data base at NIST [40]; the storage ring experiment does not comprise any wavelength measurement) is the nominally slightly stronger decay branch. However, using the N tube and a 310 nm filter of 15% transmission, the signal-to-noise ratio was rather poor, whereas the VUV transition at 233 nm (using the Q tube without any filter) yielded data with an excellent signal-to-noise ratio. For Ni^{14+}, both decay branches (208.55/281.8 nm) fall into the detection range of the Q tube; no interference filter was used. The detection efficiency of the Q tube is much lower at the longer of the two wavelengths, so that the weaker branch dominated the signal. The decay curves were easily recognized as having multiexponential character. In fits with two exponentials, we assume that the primary decay ($3s^2 3p^2\ ^1D_2$ level) and the strongest cascade ($3s^2 3p 3d\ ^3F^o_4$) are the only ones. However, it is possible that there are additional long-lived cascade levels (for example, $3s^2 3d^2\ ^3G_5$), which would be expected to be individually less populated; the lifetimes of those levels have not yet been calculated. The fit results are included in table 2.

4.3. P-like ions

In the P-like ions, two candidates for slow $3d$ cascades have been identified some time ago [41]. These $J = 9/2$ levels (of lifetimes 4 and 11 ms [42], respectively, in Fe^{11+}) decay towards the ground configuration levels via several steps (M1, E2, M2 transitions). Their population should appear in the decay curves of both $3s^2 3p^3\ ^2D^o_{3/2,5/2}$ levels (but mostly in that of the $J = 5/2$ level; the $3s 3p^2 3d^2\ J = 11/2$ level lifetimes have not yet been calculated). The UV decay curves of these levels have been recorded at TSR without filters, for P-like ions of Fe, Co and Ni, respectively ([21, 12] and this work). Under these conditions, the decay curves show three distinct exponential components, with contributions from all four $3s^2 3p^3$ level decays. Among these, the $^2P^o_{1/2,3/2}$ levels are short-lived (few milliseconds) and are rather similar to each other, so that they can be handled only approximately. The $^2D^o_{3/2}$ level has an intermediate lifetime, not much longer than those of the two slow 3d levels, and the evaluation of this decay component may be affected by this cascade contribution. The last level, $^2D^o_{5/2}$, is almost one order of magnitude longer lived and its analysis should not be affected by those cascades. However, the total decay curve has been approximated by three exponential components (the fastest representing the two $^2P^o_{1/2,3/2}$ levels together), and it is quite possible that the multiexponential analysis overall is sensitive to the relatively small contribution of these $3d$ cascades. This may be the reason why in an isoelectronic display the scatter of the experimental results appears larger (about 5%, in comparison to theoretical trends which are expected to be isoelectronically smooth) than the statistical errors (which are closer to 1 - 2%) of the fit results for individual elements. A (future) correlated analysis of the data for several elements may help to unravel the mutual dependencies. The fit

results given in table 3 represent the individual analyses for each element.

4.4. S-like ions

The spectrum of Fe^{10+} is still a challenging part of the solar spectrum, and any tool that helps to check on the validity of atomic structure calculations in support of the spectral analysis is welcome. The level structure of the ground configuration is close to that of Fe^{12+}, except for the level sequence of the ground term. The decay curves of the $^1D_2$ levels of the two species, however, differ greatly. In the S-like ion, there is a prominent multiexponential cascade tail that has been tied to slow cascades from several $3s^2 3p^3 3d$ $J = 4, 5$ levels before [21]. At the time, no actual calculations of such cascade lifetimes were available, but now they are [32]. Further (and weaker) long-lived cascades not yet calculated might arise from levels like $3s^2 3p^2 3d^2\ ^5G_6$. Overall, the order of magnitude of the apparent time constants in the cascade tail fit to the range predicted by the calculations, but the cascades are too many to analyze by multiexponential fitting. The multicomponent cascade structure, of course, also affects the determination of the primary lifetime from the data. The fit results listed in table 2 carry the error bars indicated by the fit routine. In a situation with such cascades, the strongest (and in this case fastest) fit component may be close to the level lifetime sought, but it is not possible to specify that lifetime with a meaningful 1-σ error.

5. Conclusion

The present observations forcibly change the earlier views that under the operating conditions of a heavy-ion storage ring there are just a few low-lying levels that are populated and extremely long-lived and that therefore can easily be measured with high accuracy. We now see that this may still hold true for a number of atomic systems, but that others heavily suffer from cascade population, that is, from cascades that originate from individual levels of particularly high total angular momentum that exist in some atomic systems in configurations well above the ground configuration. In certain plasmas, these levels may act as population traps, the presence of which can strongly influence the charge state balance [24, 25]. Long-lived levels way above the ground state are difficult to compute with accuracy, because a very large number of configurations has to be included in the computation. Our own (YI, JS) work in this area addresses this challenge for ions with more than two electrons in the valence shell and thus for spectra of higher complexity.

However, even if not all atomic systems feature such peculiarly long-lived levels, the combination of many individually weak cascades may contribute to systematic errors in lifetime measurements that aim at extreme accuracy, for example, when testing whether after inclusion of the aforementioned QED correction to the M1 transition operator the calculations are complete. It may be seen as ironic that our measurements, in which we find that with our technique we suffer a substantial, about 10% systematic error of

specific lifetime measurements in Al-like ions, may hold a clue as to why some other measurements that claim hundred times higher accuracy may be suffering a similar (although much smaller in absolute terms) systematic error that could possibly resolve their present disagreement with highly developed calculations, or at least reduce the disagreement by a considerable fraction (by 0.2% to 0.4% of the lifetime value). For our own MR-MP calculations, the lifetime measurements on Al-, Si-, and S-like ions corroborate once more how reliable the results of some computational approaches are, and how necessary it is to be aware of certain atomic structure details when evaluating decay curves. A systematic tabulation of such MR-MP lifetime computations for a wider set of elements is in preparation.

Because of different isoelectronic trends of primary and cascade lifetimes, in some cases it may be possible to select elements for which multiexponential analysis is more reliable than for others (see the examples of Al-like Fe and Ni). The only reliable way out of the cascade problem in atomic lifetime measurements will be by selective excitation, as has been demonstrated (without a lifetime measurement) with an electron beam ion trap at an EUV laser facility [51]. This situation reminds once more of fast-beam spectroscopy in which selective laser excitation of atoms and ions in low charge states has enabled a number of cascade-free lifetime measurements. Visible light laser excitation of ions trapped in an EBIT has been tried at Livermore, Oxford and Heidelberg, but only with very limited success. Apparently this is not a quick route to reliable lifetime measurements, and techniques like those employed here will provide useful approximations for the time being, even with some cascade contribution to recognize and to take into account.

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Tables and table captions

Table 1. Experimental conditions for atomic lifetime measurements on Al-, Si-, and S-like ions of Fe, Co, and Ni.

Element	Fe[a]	Co	Ni
Al-like ions			
Beam energy (MeV)	–	41.5	44.4
Gamma factor	–	1.0008	1.0009
Beam current (μA	–	27	17 - 22
Beam lifetime (s)	–	14	23 - 27
Wavelength of interest (nm)	530.29	435.06	360.11
Si-like ions			
Beam energy (MeV)		36.1	39.2
Gamma factor	–	1.0007	1.0008
Beam current (μA	–	22	15 - 30
Beam lifetime (s)	–	10 - 12	12.5
Wavelength of interest (nm)	257.88/338.8	233.1 / 311.0	208.55 / 281.8
P-like ions			
Beam energy (MeV)		136[c]	32
Gamma factor	–	1.0025	1.0006
Beam current (μA		120	12
Beam lifetime (s)	–	50 - 70	7
Wavelength of interest (nm)[b]	264.87/398.68	201.2/229.0	186.7 / 218.4
S-like ions			
Beam energy (MeV)		25.4	29
Gamma factor	–	1.0005	1.0006
Beam current (μA		5	18
Beam lifetime (s)	–	5 - 10	4 - 5
Wavelength of interest (nm)[b]	264.87/398.68	237.3/380.1	212.55 / 281.43

a [21, 26]
b [40]
c [12]

Table 2. Measured decay components and calculated atomic lifetimes τ (in ms) in Al-, Si-, and S-like ions of Fe, Co, and Ni. Note that the fit results do not necessarily indicate the upper level lifetime of the primary decay sought to measure (see text). Except where noted, all theory values are without the QED correction of the transition operator that would take the electron anomalous magnetic moment into account.

		Fe	Co	Ni
Al-like ions	Theory			
	$3s^2 3p\ ^2\mathrm{P}^\mathrm{o}_{3/2}$	16.6[a]	9.17 [a]	5.18 [a]
		16.5[b]	9.12[b]	5.17[b]
		16.9[c]	9.29[c]	5.26[c]
		16.62*	9.17*	5.207*
	$3s3p3d\ ^4\mathrm{F}^\mathrm{o}_{9/2}$	32.1[c]	20.6[c]	13.0[c]
		18.95*	13.18*	8.93*
	Experiment			
	Single exponential evaluation	18.0±1.2[d]	9.81±0.02*	5.9±0.01*
	Double exponential evaluation	16.2±4*	(9.7)*	5.27±0.07*
		25*	failed*	10±0.7*
Si-like ions	Theory			
	$3s^2 3p^2\ ^1\mathrm{D}_2$	6.46[e]	3.88 [e]	2.34 [e]
		9.69 [a]	4.29[a]	2.51[a]
		5.82[f]	3.49[f]	2.12[f]
		7.63[g]	–	–
		7.534*	4.366*	2.576*
	$3s^2 3p3d\ ^4\mathrm{F}^\mathrm{o}_4$	9.66[g]	6.06[g]	4.98[g]
	Experiment			
	Double exponential evaluation	8.1±0.2[d]	3.69±0.21*	2.25±0.05*
		9.9±0.4[d]	6.59±0.24*	4.85±0.17*
S-like ions	Theory			
	$3s^2 3p^4\ ^1\mathrm{D}_2$	9.83[a]	5.90[a]	3.62[a]
		9.84[h]	5.95[h]	3.64[h]
		9.80[i]	5.88 [i]	3.60[i]
		11.01[j]	6.6 [j]	3.99[j]
	$3s^2 3p^3 3d\ J=4$	1.6 - 40*	1.06 - 23.6*	0.7 - 14*
	$3s^2 3p^3 3d\ J=5$	117*	69*	40*
	Experiment			
	Multi-exponential evaluation	11.0±0.5*	6.91±0.05*	4.06±0.12*
		37	28.9±1.7*	27±0.4*
		160	183±18*	

a [49], rather similar numbers are listed in the online data basis at NIST [40]
b Semiempirical combination of transition energy and M1 line strength,
plus QED correction (EAMM) of the M1 transition operator
c FAC code [27, 38]
d [26]
e [46]
f [45]
g [29]
h [48]
i [50]
j [32]
k [21]
∗ This work

Table 3. Measured decay components and calculated atomic lifetimes τ (in ms) in P-like ions of Fe, Co, and Ni. Note that the fit results do not necessarily indicate the upper level lifetime of the primary decay sought to measure (see text).

		Fe	Co	Ni
P-like ions	Theory			
	$3s^2 3p^3\ {}^2\mathrm{D}^\circ_{3/2}$	20.8^a	11.05^a	6.13^a
		16.2^b	8.76^b	5.63^b
		16.0^c	8.61^c	4.86^c
		18.4^d	–	5.52^d
		18^e	–	–
	$3s^2 3p^3\ {}^2\mathrm{D}^\circ_{5/2}$	369^a	167^a	79.7^a
		304^b	136^b	73.6^b
		313^c	149^c	69.1^c
		314^d	–	69.8^d
		$316/323^e$	–	–
	Experiment			
	Multiexponential evaluation	18.0 ± 0.1^f	10.9 ± 0.1^g	$5.3\pm0.05^*$
		306 ± 10^f	151 ± 1^g	$70\pm1^*$

a [49], rather similar numbers are listed in the online data basis at NIST [40]
b [47]
c [43]
d [44]
e [41]
f [21]
g [12]
$*$ This work

Figure captions

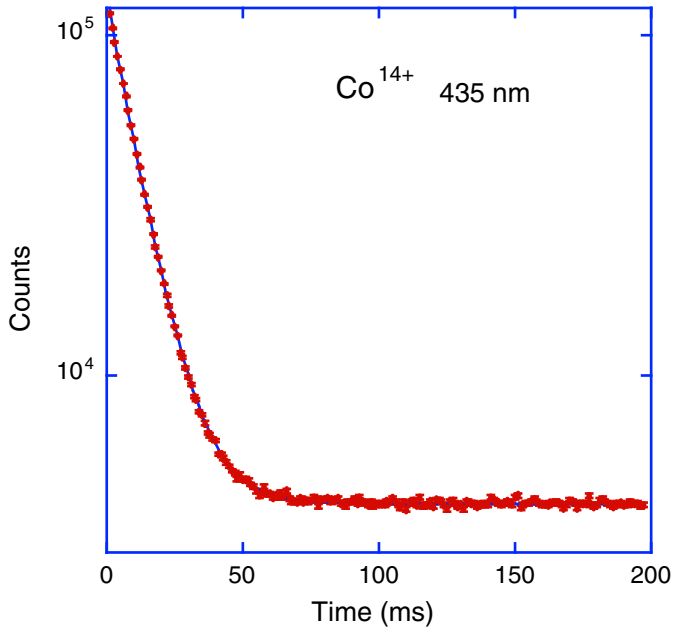

Figure 1. Sample decay curve (first two data channels covering the injection process have been deleted) and fit of a single exponential to the data (colour online). Note that the single exponential fit is excellent, but the appearance is deceptive, hiding the fact that two exponentials of not very different time constants are present. The data points bear statistical error bars. Accumulation time per channel: 164 s.

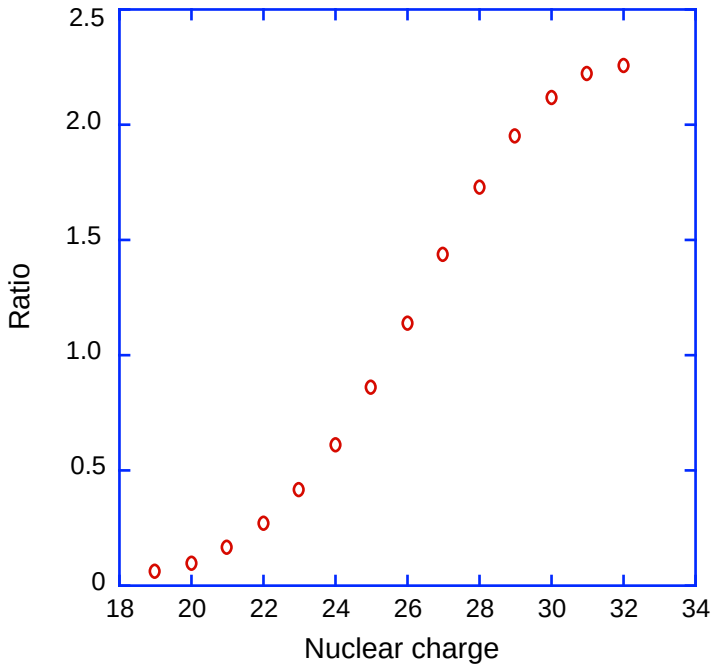

Figure 2. Ratio of (MB-MR calculated) lifetimes of the $3s3p3d\ ^4\mathrm{F}^{\mathrm{o}}_{9/2}$ and $3s^23p\ ^2\mathrm{P}^{\mathrm{o}}_{3/2}$ levels in Al-like ions of the iron group elements.