M1 and E2 decay-dependent lifetime of the lowest $^1S_0$ level in C-like ions up to Ne$^{4+}$ measured at a heavy-ion storage ring

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Abstract. Electric-dipole forbidden transitions within the ground configuration of many ion species play an important role in the diagnostics of terrestrial and astrophysical plasmas, and knowing their rates is essential for collisional-radiative models and spectral simulation. The dominant decay channel of the $2s^22p^2^1S_0$ level in the ground configuration of C-like ions changes from electric quadrupole to magnetic dipole within the first few ions of the isoelectronic sequence. Using an electron cyclotron resonance ion source at the TSR heavy-ion storage ring, we realize a lifetime measurement on the system Ne$^{4+}$ where the magnetic dipole decay channel becomes dominant. These data complement our earlier storage ring lifetime measurements on N$^+$, O$^{2+}$ and F$^{3+}$, which we discuss together with the present new result and the available theoretical calculations in their isoelectronic context.
1. Introduction

The lowest excited levels in multi-electron ions are usually those of the ground configuration. Since they are of the same parity, electric dipole (E1) transitions between them are forbidden, and only higher-order multi-pole transitions such as magnetic dipole (M1) and electric quadrupole (E2) are allowed. The transition rates are many orders of magnitude lower than those of E1 transitions involving the valence shell, and the transitions are not seen under our usual environmental or laboratory conditions. For this reason, the electric-dipole forbidden transitions, although observed in planetary nebulae since the middle of the nineteenth century [1, 2], and also in the solar corona during solar eclipses, were misinterpreted as representing hypothetical elements such as nebulium or coronium that appeared not to exist on the Earth and that spawned (in hindsight) wild ideas about their specific atomic structure. In the 1920s, Bowen became aware that such transitions of low transition rate compete with collisional excitation and de-excitation and therefore become visible only in environments of sufficiently low collision frequency, such as dilute terrestrial or astrophysical plasmas [3–5]. In many of the latter, E1-forbidden transitions may even play a dominant role. For example, when Huggins [1, 2] had built a powerful telescope expecting to see a (hoped-for) spectacularly rich spectrum of the Cat’s Eye nebula, he found just a single strong spectral line that then had to wait another 60 years for its proper interpretation as an E1-forbidden transition in O\(^{2+}\) (by Bowen). The coronium lines were not properly identified before 1942 when Edlén [6] demonstrated that they corresponded to similar E1-forbidden transitions between ground configuration levels of highly charged ions of iron group elements; this identification revolutionized our views of the conditions and processes taking place above the solar photosphere. Further references on the discovery and interpretation history of these lines have been given in [7].

In modern-day plasma diagnostics, E1-forbidden transitions play key roles. The ground configuration levels are those with the lowest excitation energies; hence any observation of the transitions within the ground configuration reveals the presence of the respective ion charge state, that is, the presence of the element and a rough estimate of the temperature necessary to reach this charge state by successive ionization effected by a more or less Maxwellian electron energy distribution. In more detail and with quantitative results, the E1-forbidden transitions compete with collisional excitation and de-excitation, and thus their transition rates are essential for collisional-radiative models of a plasma and its spectrum; synthetic spectra in comparison with the observed ones permit the determinations of density and temperature via the influence of ground configuration level populations on relative line intensities.
The electrodynamical multipole expansion of the radiation field sees M1 and E2 transitions within the same expansion order. However, in many ions, E2 transitions appear as a small contribution to M1 transitions. For example, in the transition between the fine structure levels of the $2s^22p^2$ $^2P^o$ ground term of (not too heavy) B-like ions, the M1 amplitude amounts to more than 99% of the total, and the E2 contribution to only about one-third of 1% of the same transition. The percentage changes with increasing nuclear charge $Z$, because the two transition types scale differently with the transition energy ($\Delta E$) for M1 versus ($\Delta E$) for E2 transitions), but the relative contributions are difficult to assess experimentally, if, as in this example of B-like ions, they occur at the same transition energy, where only the angular distribution of the emission is expected to show different patterns.

In order to test predictions of M1 and E2 transition rates, it is helpful to study cases that permit individual observations of the decay modes and to consider cases where the transition rates are of a similar order of magnitude. One such case is provided by the $ns^2np^2$ $^1S_0$ level in C-like and Si-like ions and similar atomic systems (see figure 1). At the beginning of the isoelectronic sequence, this level decays predominantly by E2 transition to the $ns^2np^2$ $^1D_2$ level, while in more highly ionized atoms the dominant decay is an M1 transition to the $ns^2np^2$ $^3P_1$ level. There is also a very weak E2 decay branch to the $ns^2np^2$ $^3P_2$ level. This change is illustrated in figure 2. In earlier experiments [7, 11], we addressed the C-like ions N$^+$, O$^{2+}$ and F$^{3+}$. In this paper, we report on a lifetime measurement of the $2s^22p^2$ $^1S_0$ level in the C-like ion Ne$^{4+}$ where, in comparison with F$^{3+}$ and the lighter systems, the M1 decay becomes even more dominant. In particular, from F$^{3+}$ to Ne$^{4+}$ the intensities of the M1 and E2 decay branches both show large relative changes, each of them amounting to 30–40%. The need for Ne ions required us to make a modification of the beam injection method used in these experiments (the previous work used a tandem accelerator) and to implement an electron cyclotron resonance ion source and a radiofrequency linear accelerator (RFQ) for injecting ion beams into the Heidelberg heavy-ion storage ring TSR. The present investigation is the first one to use this arrangement.

We review the context of our lifetime measurement in comparison with several theoretical treatments along the carbon isoelectronic sequence in the range of moderate nuclear charge, where the relative roles of the dominant decay contributions vary in a most pronounced way.
Figure 2. Percentages of the dominant decay branches of the $2s^22p^2{}^1S_0$ level in the first four C-like ions of the isoelectronic sequence, as predicted by various calculations [8–10]. The closely spaced results of these calculations have been averaged for this presentation. Full symbols mark the ion of present interest, Ne$^{4+}$.

2. Experiment

The level lifetime of the $2s^22p^2{}^1S_0$ level in the first four ions of the C isoelectronic sequence varies from just under 1 s in N$^+$ [7] to about 140–150 ms in the Ne$^{4+}$ ions of this work. For the measurements of the excited state lifetimes, radiative emission is observed on stored ions of the desired charge state. The storage lifetimes for these ions are chosen to be as long as possible so that their influence on the measured decay curves of the radiative emission is kept small. Hence, the decay of radiative emission is dominated by the natural lifetime of the excited levels with only small corrections due to ion loss in the storage device. An overview of long atomic lifetime measurements using ion storage rings and ion traps is given in [12, 13].

Our experiment used the TSR heavy-ion storage ring at the Max Planck Institute for Nuclear Physics, Heidelberg, Germany, combined with passive sideways observation of photon emission. A small fraction of the 55 m circumference of the ion orbit in the storage ring is observed through an optical window in the vacuum enclosure. The basic procedures have been described previously [14, 15] so we constrain this presentation to the essentials. In earlier measurements on N$^+$, O$^{2+}$ and F$^{3+}$ ions [7, 11], the ions to be stored were provided by a tandem accelerator with an internal gas stripper. However, neon does not provide negative ions, as required for injection into a tandem accelerator. A high-current injector [16] based on an electron cyclotron ion source (ECRIS) was used instead. A $^{20}$Ne$^{4+}$ beam from this source was brought to an energy of 10 MeV in an RFQ accelerator and was then further accelerated to 14 MeV in a series of radiofrequency cavities, from where it was guided into the heavy-ion storage ring. The ion beam phase space was filled by the multi-turn injection method [17] during a time corresponding to $\approx$70 revolutions in the storage ring (of about
5 μs each). After the injection the ion beam was left coasting for 500–800 ms. Although the ions travel in ultrahigh vacuum of ≈ 3 × 10^{-11} mbar, they collide with residual gas atoms and suffer (mostly) small-angle scattering and charge changing collisions. The storage time constant limited by these loss processes was measured by observing the decrease of the total stored beam current. A measurement employing the beam profile monitor system yielded 18 s. We adopt the corresponding ion loss rate of 0.06 ± 0.02 s^{-1} with an estimated uncertainty that takes into account the low ion beam current and hence low signal level.

During the time of coasting, a photomultiplier mounted on a side window of the storage ring vessel (opposite a reflective light collection system [14]) monitored the light emission from the stored ion beam; the signal counts were sorted into 1 ms wide time bins. After the observation period the ion beam was dumped and the ion trap filled again with fresh ions from the source.

The ion source produced a continuous current of Ne^{4+} ions of 500 nA. The ion species was identified by observing the charge state distribution behind the ion source. A cross check was made in the storage ring which acts as a high-quality filter for ions of a given charge-to-mass ratio, while a Schottky noise pick-up serves to establish the ion velocity with high accuracy.

After acceleration and injection in the TSR, a stored ion beam current of 800 nA was achieved, much less than what had been available in most of the experiments with a tandem accelerator injector (which cannot provide Ne ion beams, alas). This relatively low value resulted from beam current losses due to the relatively wide ECRIS emittance pattern, the RFQ micropulse structure, the limited beam transfer efficiency and from the beam current gain due to the multi-turn injection into the storage ring. Compared to the most favorable ion species in our previous optical lifetime measurements using the storage ring technique with a tandem accelerator as the injector (e.g. [14]), the stored particle current (≈ 200 nA in the present case) was lower by about two orders of magnitude.

In comparison with our standard injection scheme using a tandem accelerator, the radiofrequency acceleration scheme used in the present case also leads to a considerably larger momentum spread of the injected beams (0.5% versus fractions of 10^{-3} in the case of the tandem accelerator). Hence, the initial ion loss mechanisms in the storage ring following the injection may be different, and a significantly larger number of particles on unstable orbits may result in the initial storage period of the ion beam.

For observing optical emission from the 2s^2 2p^2 1S_0 level in Ne^{4+} ions, the favorable option is to observe the M1 decay (with a small E2 admixture) leading to the 3P term with a wavelength of ≈ 157 nm. However, radiation of this wavelength is already strongly attenuated by the sapphire window of the vacuum enclosure. We therefore resorted to observing the slightly weaker decay branch near 297 nm which leads to the 1D_2 level. An EMR 541 N-type photomultiplier tube (dark rate 25–40 counts per second) was used in single photon counting mode to observe the light from the circulating ions through either a 25 mm diameter interference filter of about 50% transmission and bandpass 10 nm around a central wavelength of 296 nm, or a 50 mm diameter filter of wider bandpass (centered at 280 nm) but lower transmission. At these near-optical wavelengths, the photomultiplier observed a background from stray light that entered the storage ring vacuum enclosure in spite of extensive precautions. The background level varied between a few hundred counts per second at daytime and 25–50 counts per second at night-time.

The optical decay curves were measured in six multi-hour data accumulation runs. Starting evaluation at about 30 ms after the injection, all runs consistently showed a slow decay of the
Figure 3. Photomultiplier signal obtained with Ne\(^{4+}\), from 30 to 800 ms after ion injection into the storage ring observing the E2 decay of the 2s\(^2\)2p\(^2\)\(^1\)S\(_0\) level near \(\lambda = 297\) nm for 61 s per channel (1 ms bin size). A single exponential (plus constant background) fit curve is indicated. From the fit to the run shown here (which represents only part of the full data), a decay time constant of \(7.7 \pm 2\) s\(^{-1}\) is derived for the decay of the optical emission before applying the storage time correction.

Optical signal with a time constant of \(\approx 130\) ms, close to the predicted lifetime of the \(^1\)S\(_0\) level. In four of the six runs, the optical decay curves also showed a fast component (\(\approx 2\) ms time constant) with a tail of \(\approx 10\) ms time constant. This initial transient was not stable during the data accumulation period and temporarily disappeared, but it reappeared when the ion beam injection into the storage ring had to be retuned. For the affected accumulation times, the first 50 ms of data storage bins were discarded before evaluation.

Sample data from the runs with a measuring time of up to 800 ms after each injection are shown in figure 3. In this figure, a weak exponential decay can be discerned on a high background. It should be noted that, in addition to the low stored ion current, the measurement is also affected by the small relative statistical weight of the investigated \(^1\)S\(_0\) initial level, which represents only 1/15 of the 2s\(^2\)2p\(^2\) ground configuration of the Ne\(^{4+}\) ion. Further measurements, in total representing about five times the accumulation time per bin as shown in figure 3, were made at a measuring time of up to 500 ms after each injection.

The data of the individual runs were fitted with single exponential decay curves plus a constant background. The average of these fit results, which scatter within 20–30\% (compatible with the data statistics), yields a decay rate of the radiative emission of \(7.85 \pm 0.9\) s\(^{-1}\). To correct for ion losses from the stored sample during the measurement, an ion beam decay rate of \(0.06 \pm 0.02\) s\(^{-1}\) determined from the ion-current decay measurement was subtracted. Finally, the decay constant was corrected by the (practically negligible) relativistic time dilation factor.
Figure 4. Predicted lifetime of the $2s^2 2p^2 \, ^1S_0$ level in C-like ions of elements N through Ne. Visualization of the scatter of all theoretical predictions cited in the three papers of this investigation ([7, 11] and this work).

of 1.0008. This result we interpret as the radiative decay constant of the $^1S_0$ level in Ne$^{4+}$, for which we thus obtain a natural lifetime of $128 \pm 16$ ms.

This is the result of an evaluation procedure unbiased by the human recognition that the majority of recent theoretical results point to a level lifetime just longer than 140 ms. However, we suspect an unintentional bias in the fit procedure that relates to the problem of observing an exponential decay curve on top of a dominant background. Some of us have recognized and discussed such a bias before, but no quantitative model has been developed so far. Hence this topic is not settled, but the bias seems at least plausible. The problem lies in the fact that the tail of the decay curve cannot reliably be discerned from the high background. No statistical test can distinguish the low-amplitude decay curve tail from statistical fluctuations of the underlying dark rate cum noise. By anecdotal evidence, the fitting process tends to return too short a decay time under such circumstances. In the present case, a bias of 10% would explain the deviation of the face value of our measurement result from the bulk of the predictions, which seems conceivable. However, without tools for a proper quantification of such a bias, we stay with the aforementioned fit results. Only a much better signal-to-noise ratio would at the same time reduce the suspected bias and, more importantly, also result in a smaller overall measurement uncertainty.

3. Discussion

The lifetime predictions for the $2s^2 2p^2 \, ^1S_0$ level in C-like ions of elements N through Ne are displayed in figure 4. The scatter of the predictions, especially for O$^{2+}$, with the spectrum
Table 1. Lifetimes (in ms) measured by the ion storage ring technique for the $2s^22p^21S_0$ level in C-like ions and selected theoretical predictions (see also the text).

| Ion  | Dominant decays, $\lambda$ (nm) | Experiment | Theory          |
|------|---------------------------------|------------|-----------------|
| N$^+$| 575                             | 910 ± 220$^a$ | 867$^d$, 898$^g$, 947$^i$, 856$^k$, 853$^l$ |
| O$^{2+}$ | 232.16/436.45     | 530 ± 25$^b$   | 517$^d$, 1269$^e$, 1333$^f$, 336$^g$, 392$^i$, 559$^j$, 523$^k$, 519$^l$ |
| F$^{3+}$ | 187.58/353.33   | 304 ± 5$^b$     | 294$^d$, 608$^g$, 199$^h$, 299$^i$, 312$^k$, 300$^l$, 312$^m$, 295$^n$ |
| Ne$^{4+}$ | 157.475/297.313 | 128 ± 16$^c$   | 142$^d$, 170$^e$, 101$^f$, 170$^h$, 147$^i$, 146$^j$, 144$^k$, 147$^l$, 142$^m$ |

$^a$ Träbert et al [7].
$^b$ Träbert et al [11].
$^c$ This work.
$^d$ Nussbaumer and Rusca [8].
$^e$ Cheng et al [18].
$^f$ Bhatia et al [19].
$^g$ Kaufman and Sugar [22].
$^h$ Bhatia and Doschek [20].
$^i$ Vilkas et al [23].
$^j$ Galavis et al [9].
$^k$ Froese Fischer and Tachiev [10].
$^l$ NIST on-line database [24].
$^m$ Jönsson et al 2011 [25].

(O III) of highest astrophysical interest, is sizeable. The highest predictions for O$^{2+}$ exceed the lowest by almost a factor of four, for F$^{3+}$ by a factor of about three, and for Ne$^{4+}$ by a factor of about two. Apparently, the various calculations converge for higher ionic charges, but the principal physics interest is in low charge states, for which there are the maximum number of astrophysical observations and the largest relative variations of the E1-forbidden decay modes.

The data entries in figure 4 are intentionally left unlabeled (but all references are cumulatively found in the three papers of this investigation, namely [7, 11] and this work), demonstrating the situation of an experimenter trusting everybody’s calculations similarly. Some filtering is useful in improving the predictive value of any such calculations. To illustrate this point, the two highest lifetime values predicted for O$^{2+}$ are higher than all predictions for N$^+$ (which is in conflict with any isoelectronic scaling trend). One of the two calculations is a multi-configuration Dirac–Fock (MCDF) computation by Cheng et al [18] that may not be expected to be really applicable to such low-charge-state ions (and does much better for high-Z ions), whereas the other is a superstructure code calculation by Bhatia et al targeted at O$^{2+}$ and apparently not doing better; in fact, the result is farther away from the bulk of the other predictions and from experiment than even the ill-suited MCDF survey calculation. A corresponding calculation by Bhatia and Doschek [20] treats Ne$^{4+}$ and yields the longest lifetime prediction there, too. For further discussion we concentrate on a comparison of the experimental data with only those calculations that seem better suited than others and that cover several elements of the sequence, because our interest is in the isoelectronic trends.

The experimental lifetime results for the $2s^22p^21S_0$ level in C-like ions of elements N through Ne are given in table 1, along with the results of various predictions. A selective
Figure 5. Lifetime of the $2s^22p^2 \, ^1S_0$ level in C-like ions of elements N through Ne. Experimental results and predictions by those theoretical studies from figure 4 that cover several ions of the sequence, but without calculations that are deemed overstretched for low-charge-state ions. Evidently, the scatter of the predictions in this sample is much reduced by this selection. Experimental results ([7, 11] and this work) are shown by full squares with error bars. The theoretical work (see the legend) is identified by the lead authors in table 1.

Isoelectronic comparison that emphasizes the isoelectronic trend (figure 5) shows a number of calculations in a band that encompasses the experimental findings. Nevertheless, there are calculations that agree with experiment for one or two elements and clearly disagree for others. The comparison with experiment and other theory indicates that in these cases the shortcomings are with those individual calculations.

Overall, the present result on $Z = 10$ clearly confirms the trend that recent calculations describe the transition rates of the lower-$Z$ ions rather well. In particular, experiments have now probed a range of $Z$ in which the relative strengths of the two transition types E2 and M1 involved in the $^1S_0$ level decay are of comparable size, but vary significantly. The good agreement noted previously for F$^{3+}$ extends also to the present result and thus confirms that both transition types are correctly treated by the recent computations.

Unfortunately, the accuracy of the present experimental result is not sufficient for a detailed assessment of the small deviations among the recent predictions for Ne$^{4+}$. What are the prospects for better measurements beyond perhaps a factor-of-two improvement of the ion current from the source, which might improve the statistical reliability of the data, but would not significantly do so? As discussed elsewhere [21], atomic lifetime measurements have optimum working ranges in terms of atomic lifetime and detector sensitivity. The lifetimes of the $2s^22p^2 \, ^1S_0$
level in C-like ions of elements N through Ne are on the long side of the optimum range. Regarding sensitivity, all four ions suffer from the same low statistical weight of a $J = 0$ level and thus a low-level population and decay signal. However, the wavelengths of radiative emission accessible for observation also play an important role. In O$^{2+}$ and F$^{3+}$ ions, one of the decay branches falls into the operating range of solar blind detectors with their excellent signal-to-noise performance; for this reason the experimental results for these two ion species are much more precise than for the other two. For Ne$^{4+}$, the wavelength of this decay branch becomes too short for transmission through optical windows. Therefore the decay branch in the near-visible spectral range, leading to the $^1\text{D}_2$ level, has to be selected instead, with corresponding detrimental effects of stray light background. For Na$^{5+}$ and Mg$^{6+}$, this decay branch falls into the detection range of solar blind photomultipliers. These ion species, however, can only be provided after modifications of the TSR ion source technology that are currently not within reach. Moreover, all measurements towards heavier systems beyond Ne$^{4+}$ have to face the decrease with $Z$ of the relative yield of the M1 decay branch leading to the $^1\text{D}_2$ level (see figure 2). Hence, the properties of this atomic system limit the achievable experimental accuracy in comparison with other recent atomic lifetime studies. Nevertheless, the sensitivity achieved is sufficient to clarify inconsistencies among existing theoretical predictions and is considerably smaller than the relative intensity changes of the E2 and M1 transition branches.

4. Conclusions

On the technical side, this is the first experiment to make use of an ECRIS for multi-turn injection into the TSR heavy-ion storage ring. This source type has demonstrated the provision of a rare-gas ion beam that was not available from the regularly used tandem accelerator injector. In measurements that are less photon-starved than the present one, such an extension of the elemental coverage will be most welcome. However, the emissivity of a tandem accelerator is much superior to the rather wide emission profile available from this ECRIS, and this mismatch severely limits the storable ion beam current and thus the photon signal rate. A second hurdle was not the pulse structure of the RFQ accelerator between the ion source and the accelerator (the high-current injector), but the minimum charge-to-mass ratio (>1/9) this accelerator needs in order to work properly. The ratio excludes many low-charge-state ion species of the rare gases that would be of astrophysical interest.

On the scientific side, the source output turned out to be only barely sufficient to add a meaningful data point to a systematic study of C-like ions that has spanned more than a decade. In the course of this investigation, the lifetime of the $2s^22p^2\,^1\text{S}_0$ level in C-like ions of elements N through Ne has been measured and thus the variation from a level lifetime fully dominated by the E2 radiative decay channel (in N$^+$) to one with a majority M1 decay channel (in Ne$^{2+}$) documented and quantified. Few atomic systems yield as good access to M1 versus E2 dominated level decays as do C-like ions. The measurements identify a few calculations that match the experimental findings for the isoelectronic sequence and that therefore qualify for future usage in radiative-collisional plasma modeling. Of course, this benchmarking points to other calculations being less suitable.

The present investigation is part of an evolving web of such measurements that employ various types of ion traps (conventional traps, heavy-ion storage ring, electron beam ion trap and so on; see [12, 13, 21]). Some spectacularly accurate lifetime measurements (with uncertainties of a small fraction of 1%) have been reported, which test the accuracy of the quantum
mechanical treatment of few-electron systems and possibly of leading QED corrections. There are still some suspected sources of systematic error that need to be investigated so that the full potential of the techniques can be exploited. The majority of the data carry uncertainties of one to a few per cent, which is very good and certainly much better than the reliability of most predictions. Of course there are also measurements such as the present one which are of lower accuracy, because they challenge the performance envelope of such experiments at the edge of the practical parameter space. Yet such measurements are helpful in completing the picture, ascertaining isoelectronic trends and providing a challenge to find ways of further improvement.

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