Improvement in the reliability of lifetime measurements for highly charged ions in the 0.01–100-ns range

Y. Zou,* R. Hutton, S. Huldt, and I. Martinson
Department of Physics, University of Lund, S 223 63 Lund, Sweden

K. Ando, T. Kambara, H. Oyama, and Y. Awaya
Atomic Physics Laboratory, RIKEN, Wako shi, Saitama 351 01, Japan

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We report here the finding that, to obtain reliable lifetimes in highly ionized atoms measured by beam foil excitation, the conditions must be set so that the charge state studied is well below the average charge of the foil emergent beam. If this condition is not fulfilled, the decay curves may be significantly distorted by blending with close-lying satellite lines. We present evidence to show that failures to follow this criterion have led to serious underestimates in reported lifetimes in isoelectronic line strength studies. We also suggest guidelines for avoiding these distortions in future measurements. [S1050-2947(99)03308-9]

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Reliable experimental values of lifetimes and transition probabilities for highly charged ions provide important tests of atomic many body theories in the relativistic regime. Furthermore, such data have applications in a number of fields, including astrophysical studies of the solar corona, solar flares and the atmospheres of hot stars, as well as the diagnostics of fusion plasma impurities.

Until some years ago, the beam-foil method was the only experimental technique for such measurements [1,2]. More recently very valuable results have been obtained by using other methods, e.g., heavy-ion storage rings [3,4], or electron-beam ion traps [5,6]. In this way very long lifetimes (ms) but also very short (fs) lifetimes [6] can be measured. These techniques nicely complement the beam-foil method which can be applied in the 0.01–100-ns interval where many atomic and ionic lifetimes fall.

However, a systematic shortening of the experimentally obtained $3s3p\ ^3P_1$ lifetimes of Mg-like ions, in comparing with theoretical predictions, was noted in our recent beam-foil studies of the isoelectronic $3s^2\ ^1S_0$–$3s3p\ ^3P_1$ intercombination line strengths [7]. This discrepancy appeared to increase with the charge of the ion, being as high as 29% for Mg-like Nb^{29+}. Since the relativistic atomic structure theories become more reliable as Z increases, no good explanation could be given of this discrepancy.

Similar lifetime shortenings have been noted in some earlier beam-foil works, but no systematic studies have been reported. In a pioneering study of the lifetime of the $1s2s\ ^3S_1$ level in He-like Ar^{16+} which decays by an $M1$ transition to the $1s^2\ ^1S_0$ ground state, Marrus and Schmieder [8] obtained a lifetime which was about 20% shorter than the theoretical value. In another beam-foil study, for Cl^{15+}, Cocke *et al.* [9] found a similar discrepancy between theory and experiment. Following these and some other experimental results, which, e.g., noticed that the decay of $1s2s\ ^3S_1$ was nonexponential [10], Lin and Armstrong [11] proposed an explanation of these discrepancies. They assumed that the $1s^2\ ^1S_0$-$1s2s\ ^3S_1$ $M1$ transitions in beam-foil spectra were contaminated by unresolved Li-like satellites $(1s^2\ ^1S_0)nl$-$(1s2s\ ^3S_1)nl$, these were found to decay faster than $M1$ transitions in He-like ions.

If the shortening of the experimental decay times is from the satellites, we should be able to solve this problem by increasing the ion beam energy, as at higher energy the charge state with one more electron, which leads to the satellites, will be less abundant. In the present experiment we measured the decays of the $1s^22s\ ^2S_{1/2}$-$1s^22p\ ^2P_{1/2,3/2}$ transitions in Li-like Mg^{9+} at different beam energies, to study systematically the beam energy dependence of experimentally obtained decay times of $1s^22p\ ^2P_{1/2,3/2}$ levels. Considering that the unresolved satellites would have different contribution to the different part on the measured line profile, we also studied the wavelength dependence of the decay time for the $1s^22s\ ^2S_{1/2}$-$1s^22p\ ^2P_{3/2}$ transition in Li-like Ar^{15+}. In these cases there can be no reasonable doubt that the theoretical lifetimes for the excited levels are very accurate [12,13], whereas somewhat larger theoretical uncertainties are expected for Mg-like intercombination lines studied in Ref. [7].

The experiments were carried out using the beam-foil setup at the RILAC linear heavy-ion accelerator at RIKEN. This setup has been previously described in Refs. [14,15]. For the present experiments the conditions have been improved by the addition of a CCD detector system. This system has two important advantages which were utilized in the experiments: (a) the ability to record two-dimensional images and (b) the noise reduction capabilities by using the photon energy resolution of the CCD chip.

The spectral lines of interest were isolated using a 2.2 m grazing incidence spectrometer. The wavelengths of the resonance doublet in both Mg^{9+} and Ar^{15+} are well known (609.80 and 624.95 Å for Mg, 353.87 and 389.08 Å for Ar) [13] and these lines are well separated spectroscopic features. An example of a recorded spectrum for the Ar $^2S_{1/2}$–

*Present address: Applied Physics Department, Shanghai Jiao Tong University, Shanghai 200030, P.R. China.

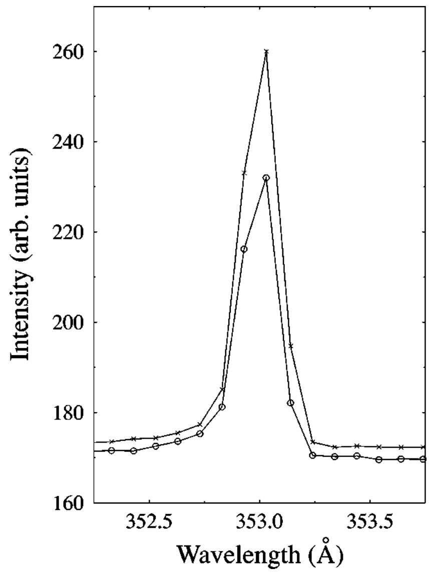

FIG. 1. The $1s^22s\,^2S_{1/2}$–$1s^22p\,^2P_{3/2}$ line in Ar^{15+} recorded at 0.06 and 0.24 ns after excitation. In this case the 1200-lines/mm grating was used. The points within the FWHM were determined by the CCD pixel size and the required resolution.

$^2P_{3/2}$ line is depicted in Fig. 1 (raw data), which shows a certain asymmetry which is more pronounced on the short wavelength side. These lines were then studied as a function of time after excitation by recording CCD images at different downstream foil-spectrometer entrance slit separations (referred to as the foil-slit separation later in the text). In the Mg studies a 600 lines/mm grating was used whereas both this grating and a 1200 lines/mm grating were used in the Ar case. With the latter grating an improved spectral resolution was obtained.

In the case of Mg, three beam energies were selected, 0.4, 1.3, and 2.5 MeV/u, the foil thickness here was 10 $\mu g/cm^2$. These energies were chosen to provide pronounced differences in the charge state distribution after the ion-foil interaction [16]; see Table I.

The intensity decay curves were generated by integrating the counts under the whole line profile for the different foil-slit separations. Each decay curve was analyzed using a multiexponential fitting routine and it was found that a two exponential solution was required to fit the experimental data. Table II lists the obtained decay times (with relative uncertainties around 10%) together with recent theoretical results [12,13]. A weak but long decay component has been found in all cases in fitting. We removed the influence of this long decay (for illustrative purpose) and show in Fig. 2 the decay curves of the $^2S_{1/2}$-$^2P_{3/2}$ transition in Mg^{9+} recorded at the three energies.

TABLE I. The equilibrium charge-state distribution for Li- and Be-like Mg ions after stripping by a carbon foil at the energies of 2.5, 1.3, and 0.4 MeV/u, and that of Ar at 2.6 MeV/u.

Ion	E (MeV/u)	Be-like percent	Li-like percent	Li:Be ratio
	2.5	0.2	8	40
Mg	1.3	12	40	3.3
	0.4	20	4	0.2
Ar	2.6	26	34	1.3

TABLE II. Decay times for the $1s^22p\,^2P_{1/2,3/2}$ levels of Li-like Mg ions measured at beam energies of 2.5, 1.3, and 0.4 MeV/u, $\tau_{2.5}$, $\tau_{1.3}$, and $\tau_{0.4}$ together with theoretical decay times τ_{th} from (a) Ref. [12] and (b) Ref. [13].

Level	τ_{th} ns		$\tau_{2.5}$ ns	$\tau_{1.3}$ ns	$\tau_{0.4}$ ns
$^2P_{3/2}$	1.331[a]	1.335[b]	1.33	1.15	0.61
$^2P_{1/2}$	1.438[a]	1.440[b]	1.47	1.21	0.83

As can be seen in Fig. 2, the three slopes are quite different. The same trend was observed for the $^2S_{1/2}$-$^2P_{1/2}$ line in Mg^{9+}. The decay curves thus yield different decay times for the energies used. However, at the highest energy, 2.5 MeV/u, the experimental decay times are in excellent agreement with the theoretical results; see Table II.

Applying the same model as proposed by Lin and Armstrong [11], we must consider the satellites in the Be-like system, i.e., $1s^22snl$-$1s^22pnl$ transitions. For low values of n, such lines would be outside the measured line profiles of the Li-like transitions. However, with increasing n, the satellite lines will have wavelengths very similar to those of the resonance lines in the Li-like ions.

The rates of decay for $2snl$-$2pnl$ transitions, for the values of n which contribute to unresolvable satellites, are rather similar to those of the $2s$-$2p$ lines in Li-like ions. However, the $2pnl$ levels have other decay branches due to the decay of the nl electron, i.e., $2pn'l'$-$2pnl$ decays. The result of these additional decay modes is that the $2pnl$ levels usually have shorter decay times than the $2p$ levels in Li-like ions, if

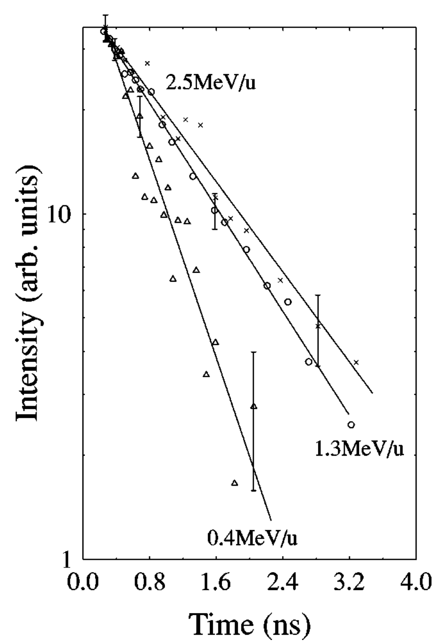

FIG. 2. Deacy curves for the $1s^22s\,^2S_{1/2}$–$1s^22p\,^2P_{3/2}$ transition in Mg^{9+} recorded at three different beam energies, 2.5, 1.3, and 0.4 MeV/u. The lifetime obtained from the 2.5 MeV/u measurement agrees with the theoretical lifetime for the $1s^22p\,^2P_{3/2}$ level.

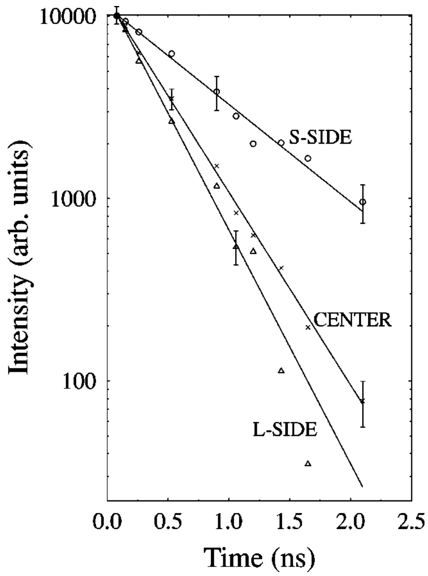

FIG. 3. The decay curves for the transition $1s^22s\,^2S_{1/2}$–$1s^22p\,^2P_{3/2}$ in Ar^{15+} generated by integrating the counts over the long- wavelength side, center part, and short-wavelength side of the measured line profile.

there are not too many cascades from higher satellite levels. At 2.5 MeV/u, the fraction of Be-like ions is small compared to the fraction of Li-like ions, see Table I, the satellite contamination will therefore be low and the measured decay times would agree with the theoretical lifetime values. As the beam energy is decreased the satellite contribution increases which should lead to decay times disagreeing with the theoretical results. This is illustrated in Table II. At 0.4 MeV/u Mg^{8+} ions are five times more abundant in the ion beam than the Mg^{9+} ions, so the experimental results are far from those of theory.

In the Ar studies, only one energy, 2.6 MeV/u, was used. Here the foil thickness was 40 $\mu g/cm^2$. The measurements were done with different spectral resolutions, where the geometry of the spectrometer allowed us to use two different gratings (600 and 1200 lines/mm). This was not possible in the Mg case. The better resolution (with line width 0.2 Å) afforded by the 1200-lines/mm grating favored studies of the variation in decay time of the different sections on the measured line profile. In this case, we studied only the decay of the stronger component of the doublet, namely the $^2S_{1/2}$–$^2P_{3/2}$ line.

We extracted the decay times from decay curves generated by integrating the counts under the whole line profile measured using both gratings, and by integrating the counts over three channels centered 0.1 Å away from the peak position at long wavelength side, over three channels centered at peak position, and over three channels 0.2 Å away from the peak at short wavelength side of the line profile. The latter three decay curves are shown in Fig. 3 together with their fits. The results are listed in Table III (with relative uncertainties around 10% for τ_w and τ_c, around 15% for τ_l and τ_s), in which theoretical lifetimes are also listed [12,13].

The decay times extracted using the whole line profiles, recorded with both gratings, are in good agreement with each other but shorter than the theoretical results by about 35%. This corroborates the explanation given above, i.e., that the

TABLE III. Decay times (in ns) of the Li-like Ar $1s^22p\,^2P_{3/2}$ level, obtained by the decays of the whole line profile recorded by the 600 line/mm (τ_w^a) and 1200 line/mm (τ_w^b) gratings, and obtained by the decays of long-wavelength (τ_l), center (τ_c), and short-wavelength (τ_s) part of the line profile recorded by the 1200-line/mm grating are listed together with the theoretical lifetimes (τ_{th}) from (a) Ref. [12] and (b) Ref. [13].

τ_{th}		τ_w^a	τ_w^b	τ_l	τ_c	τ_s
0.652[a]	0.653[b]	0.45	0.42	0.34	0.41	0.81

shortening of the decay times is caused by satellite contamination of the decay curve. At 2.6 MeV/u the Ar beam has similar abundance for Li-like and Be-like ions and therefore the satellite contamination should be quite severe. The variation of the decay time with the wavelength provides additional support to the existance of unresolved satellites lines, from $2pnl$ levels in Be-like Ar^{14+}, in the Ar^{15+} resonance line. In some recent work by Ishii *et al.* [17] satellite lines have also been discussed for beam-foil spectra recorded in the visible region.

From the discussion of Rydberg state populations of beam-foil excitation in Ref. [18], one broad maximum of level population around $n=10$ for Mg^{8+} and one around $n=16$ for Ar^{14+} would be expected. We observed $2p5g$-$2p6h$, $2p4f$-$2p5g$, and $2p3d$-$2p4f$ satellite electron transitions of Be-like Mg ions at 0.4 MeV/u, however no estimate of relative level populations can be made, as the spectrometer is not intensity calibrated. At beam energy of 2.5 MeV/u, the above transitions were not observed. The transitions involving higher n satellite electrons are beyond our spectrometer detection region.

We can note here that the satellite levels with very high n may have less influence on the measured decay curves as most of them will have an autoionization channel and hence decay mostly by rapid electron emission. Only the levels which are parity-forbidden for autoionization (with $l=L$)

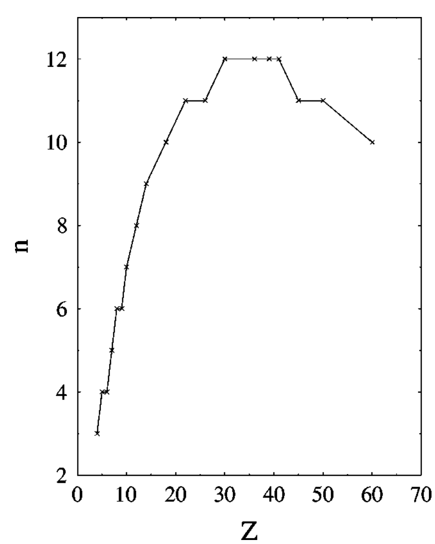

FIG. 4. The value of n in $1s^22pnl$ for which autoionization becomes energetically allowed as a function of Z for Be-like ions. The value of n is obtained from calculations using the Cowan computer code.

will keep on influencing the decay curve measurement. In Fig. 4 we show the value of n for which the $2pnl$ levels become autoionizing for ions in the Be I sequence, calculated by using the superposition of configurations (SOC) code by Cowan [19]. This calculation agrees with the more detailed investigation by Mancini and Safronova [20] who used the multiconfiguration Dirac-Fock (MCDF) code. It is interesting to note that the satellite levels, $2pnl$, become autoionizing for quite low n in low Z systems [21] and hence decay time measurements for low Z Li-like ions should have less problems than the cases discussed above. For example, most of the $2p5l$ levels in Be-like O are autoionizing and satellites from $n=4$ levels ($2p4l$) will lie far from the primary line. Therefore in such cases one would expect more reliable lifetimes by beam-foil measurements.

In reviewing the previous beam-foil work where disagreements were found between experimental and theoretical results we found that the experiments were always conducted with undesirable charge state distributions. For example, in the study of Mg-like ions [7], the highest ratio of Mg-like to Al-like ions abundance was about 6/5 [16], while in the early investigations of He-like Cl [9,10] the ratio of He-like to Li-like ions was around 1/5 [16], so the satellite contaminations were serious in all these cases.

As a summary, we investigated the reliability of the beam-foil method for lifetime (decay time) measurements in highly charged ions by systematically studying the decay curves for the resonance lines in Li-like Mg^{9+} at different beam energies. We found that decay times extracted from the measurements at the most probable charge states can be quite questionable and that different results are obtained depending on which part of the line profile is used. This observation was supported by our higher resolution measurements for Li-like Ar^{15+}. We conclude that such effects are caused by satellite contamination from ions in the beam with one extra electron. However we also found that the lifetimes measured at very high energy, when Li/Be like ratio of ions in the beam is about 40, agreed well with the theoretical lifetimes for the $1s^2 2p\ ^2P$ levels of Mg^{9+}. We assume that a systematic Z-dependent disagreement between experiment and theory concerning intercombination transition rates in Mg-like ions [7] is also due to satellite contamination because unfavorable energies were used. The reliability can be improved by performing measurements at beam energies which minimize the satellite producing charge state.

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