

**$M1$  transition rate in  $\text{Cl}^{12+}$  from an electron-beam ion trap and heavy-ion storage ring**E. Träbert,<sup>1,2,\*</sup> P. Beiersdorfer,<sup>2</sup> G. Gwinner,<sup>3</sup> E. H. Pinnington,<sup>4</sup> and A. Wolf<sup>3</sup><sup>1</sup>*Fakultät für Physik und Astronomie, Ruhr-Universität Bochum, D-44780 Bochum, Germany*<sup>2</sup>*Division of Physics and Advanced Technologies, Lawrence Livermore National Laboratory, Livermore, California 94550-9234*<sup>3</sup>*Max-Planck-Institut für Kernphysik, D-69117 Heidelberg, Germany*<sup>4</sup>*Department of Physics, University of Alberta, Edmonton, Canada AB T6G 2J1*

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The transition probability of the magnetic dipole ( $M1$ ) transition  $2s^22p^2\ ^2P^o\ J=1/2$  to  $J'=3/2$  ( $\lambda = 574.19$  nm) in the B-like ion  $\text{Cl}^{12+}$  has been measured using two different light sources, the Heidelberg heavy-ion storage ring TSR and the Livermore electron-beam ion trap EBIT-I. Our results for the atomic level lifetime are  $21.2 \pm 0.6$  ms from the heavy-ion storage ring and  $21.0 \pm 0.5$  ms from the Livermore electron-beam ion trap. Particular attention has been paid to systematic errors, making this experiment a common reference for atomic level lifetime measurements in the visible spectrum.

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**I. INTRODUCTION**

Electric dipole transitions between the fine-structure levels of a given term are forbidden because of the parity selection rule. The lowest multipole order radiations permitted are magnetic dipole ( $M1$ ) and electric quadrupole ( $E2$ ) transitions, and such transitions between the fine-structure levels of multiply charged ions are the origin of many of the solar coronal lines and are also of great interest for plasma diagnostics [1]. Their transition rates are used to determine species densities in astrophysical plasmas and are needed in the modeling of the plasma edge in fusion test devices. B- and F-like ions feature just a single such line each, corresponding to the transitions  $2s^22p^2\ ^2P^o_{1/2} - ^2P^o_{3/2}$  and  $2s^22p^5\ ^2P^o_{3/2} - ^2P^o_{1/2}$ , respectively, within the ground term. Since B-like ions are the simplest ions with such a fine-structure transition in the ground term, we will concentrate on these here. The electric quadrupole ( $E2$ ) contribution to the decay amplitude is lower than the magnetic dipole ( $M1$ ) amplitude by more than four orders of magnitude, and we therefore disregard the  $E2$  contribution.

For  $M1$  and  $E2$  transitions, the transition probability depends mostly on angular coupling factors and the energy interval. However, recent experimental transition rate data from various ion traps for argon ( $Z=18$ ), an element with prominent coronal lines from B- and F-like ions, do not all agree with each other [2–4]. Only the results obtained at the Livermore electron-beam ion trap EBIT-II are compatible with theory. Livermore data on a neighboring element, potassium ( $Z=19$ ) [5], are also available, as well as results from lifetime measurements on titanium ( $Z=22$ ) [6], obtained at the Heidelberg heavy-ion storage ring TSR. In order to gain a better understanding of the systematic errors involved, it would be good to be able to study a reference transition in the same element with both devices. This is of particular interest for atomic lifetimes of many milliseconds, since this is a range close to what has been perceived as the upper limit for any such lifetime studies using electron-beam

ion traps [7]. With the Livermore electron-beam ion trap, gas injection works best (hence Ar was studied first), and potassium could be injected as a vapor. The present injector for the Heidelberg storage ring, by contrast, is a tandem accelerator that requires negative ions; this precludes the use of Ar and would not yield sufficient ion beam currents of potassium ions. Therefore we elected to use chlorine ( $Z=17$ ), which can be produced as a strong ion beam for tandem accelerators and which can also be bled into the electron-beam ion trap from compounds with a high vapor pressure. Moreover, chlorine has been a diagnostic in magnetic fusion plasmas [8], and its forbidden lines have been well established as constituents of the coronal emission of the sun [9].

By isoelectronic comparison, laboratory data on several ions help to clarify the experimental situation, and they assist in assessing the validity of the laboratory data obtained for the elements of primary interest. Three  $M1$  transitions have been studied in Ar and K. Two of these were in B- and F-like ions, while the third one was an  $M1$  transition in an excited configuration of the Be-like ion. Of these three, only one, the  $M1$  transition in the B-like ion  $\text{Cl}^{12+}$  ( $\text{Cl XIII}$ ), has a wavelength (574.19 nm) [10,11] that is compatible with the sensitivity range of our detectors. The expected level lifetime of about 21 ms is longer than that in argon by a factor of 2, which is a challenge to the experimental techniques at the electron-beam ion trap. In fact, it is the longest radiative lifetime studied on an electron-beam ion trap so far. For the heavy-ion storage ring, such a lifetime is in an optimum range. We therefore undertook to learn about systematic errors that affect lifetime measurements with an electron-beam ion trap when using detectors for visible light.

**II. EXPERIMENT****A. Heavy-ion storage ring**

The measurements at the Heidelberg Max Planck Institute of Nuclear Physics TestSpeicherRing TSR followed the pattern established previously [12,13]. Negative chlorine ions were produced from an  $\text{AgCl}_2$ -Ag mixture in a sputter ion source, injected into a tandem accelerator, stripped to positive charge states  $q^+$  by being passed through a dilute gas in the accelerator high-voltage terminal and accelerated further

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to an energy of about 45 MeV. At the exit of the accelerator, the ions were stripped again, this time in a thin carbon foil. Magnetic fields then selected ions of the desired charge state  $q = 12^+$  (ion energy 44.3 MeV), and only ions of this single charge state and well-defined energy were then guided to the heavy-ion storage ring, some 100 m away.

A beam current of about  $5 \mu\text{A}$  was available for injection into the storage ring, where multiturn injection and the stacking technique [14] raised the ion current in the ring up to about  $100 \mu\text{A}$ . The ion source was operated in a pulsed mode. Each of the pulses was long enough (about 1 ms) to fill the storage ring with a continuous ion beam accumulated over some 30 revolutions. At a ring circumference of 55 m, the stored-beam section has a total length of  $\approx 1.7$  km. After the multiturn injection, the ions were left coasting until being ejected in preparation for the next storage cycle.

A fraction of the ions existed in collisionally excited levels from the ion-foil interaction in the stripper. The ion travel from the source to the ring required approximately  $6 \mu\text{s}$ , which is much shorter than the level lifetime of interest. This travel time was long enough, though, to let all higher-lying excited states decay to the two levels of the ground state, one of which is the level of present interest, and to thus increase their population over what was already reached in the production process. Moreover, very high-lying excited states would be quenched in the motional electric fields of the beam steering magnets.

The circulating ion cloud was stored for about 0.2 s, or about ten predicted lifetimes of the level of interest, so that the optical fluorescence signal could be observed and followed well into the background. A clock pulse, preceding the injection phase by about 1 ms, gated on each photon detection period. Photon counts were accumulated into 200 sequential channels, each 1 ms wide. The resulting data files provided a record of the fluorescence intensity emitted by the stored-ion cloud as a function of time, that is, a photon signal decay curve.

A bialkali-cathode photomultiplier tube (PMT, model EMR 541N, with a short-wavelength cutoff near 300 nm) was employed for the optical observations, in combination with the same interference filter that was used for the Livermore EBIT-I experiments (central wavelength near 570 nm, bandpass 50 nm). Since the transition of interest is the only one from the only long-lived level of the single-charge-state ions stored, the filter is not so much needed to discriminate against other spectral lines of Cl (none are expected), but only against any ambient light. In fact, the hot vacuum gauges in the nearby sections of the storage ring had to be switched off, because their stray light would be noted in the PMT signal.

The storage-ring technique has excellent charge-state selectivity and employs background pressures on the order of  $5 \times 10^{-11}$  mbars. This combination yields an optically clean environment with ions of a well-defined charge state exclusively stored in the ring. Nevertheless, there are ion losses by collisions with the rest gas. Because of the high ion velocity, charge-exchange collisions may be expected to be more frequent in the storage ring than in the electron-beam ion trap, yet the confinement is regularly higher in the former device,

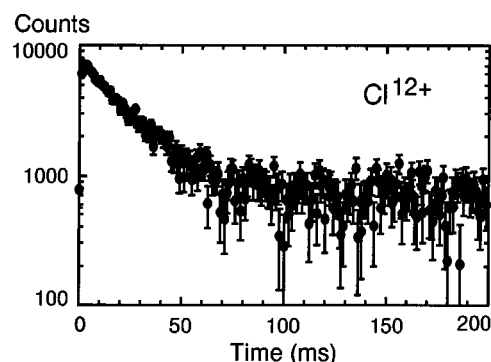


FIG. 1. Photon signal (logarithmic scale) obtained with Cl XIII at the heavy-ion storage ring. A background contribution of 45 000 counts per channel has been subtracted from the data.

despite comparable rest gas pressures. However, the charge-exchange cross sections decrease rapidly at the higher velocities (MeV ion energies) [15–17]. This decrease more than compensates the increase in the velocity, resulting in a rate coefficient  $\langle\sigma v\rangle$  that is smaller in the storage ring than in the ion trap. The ion beam current was monitored on-line by a beam profile monitor that senses the ions produced in the rest gas. The ion storage time constant was then used to correct the apparent time constant of the optical decay.

Within the coasting period of the 0.2 s cycle time, the beam current changes very little. The ion storage time constant was calculated, based on Nikolaev's early work [18], as about 46 s. Such estimates have agreed with measurements on comparable ions in earlier experiments. A fit to the beam profile monitor signal (over a time range of 200 ms) yields a beam lifetime of order 17 s, corroborating the calculational estimate. Such a long storage time in relation to the optical lifetime of about 20 ms implies a correction by 0.1% or less. Relativistic time dilation ( $\gamma = 1.00136$ ) requires a similarly small, but opposite, correction, the sum of which is presently negligible.

There are not many systematic variations of the experimental conditions at the heavy-ion storage ring that one can check for their influence on the results [13]. Since in the present case the correction for the ion storage time amounts to less than 0.1%, it seemed unnecessary to do further tests on this correction.

The largest uncertainties are those associated with the data evaluation. The signal-to-background ratio varies with the stored-ion beam current and with different settings of amplifiers and discriminator thresholds. Out of a total data accumulation time exceeding 4000 min, data records of poorer run conditions (some 10% of the total) were afterwards excluded from the evaluation. Thus data from more than  $10^6$  injections remained, or an accumulation time per data channel of about 17.6 min. The full data sample is shown in Fig. 1. Truncating data curves in the beginning (after injection, for a possible settling-down period), for times up to 20 ms, or truncating the curve tail (by up to 50 ms) for artifacts of the fit procedure, had no noticeable effect on the level lifetime obtained from a single-component exponential fit. The individual fit results remained within the statistical uncertainty of the typical value. This uncertainty,

however, is dominated by the dark rate of the photomultiplier of about 45 counts per second. We obtain as the heavy-ion storage ring lifetime result for the  $\text{Cl XIII } 2s2p^2P_{3/2}^o$  level a value of  $21.2 \pm 0.6$  ms (transition rate  $47.2 \pm 1.3 \text{ s}^{-1}$ ).

### B. Electron-beam ion trap

The measurements were carried out at the University of California Lawrence Livermore National Laboratory, using the electron-beam ion trap EBIT-I. This is the original (first ever) electron-beam ion trap [19] that later was converted into SuperEBIT [20]. At present, it is operating again in the low-energy EBIT-I mode ( $\leq 20$  keV). Like SuperEBIT [21], EBIT-I is capable of running at electron-beam energies well below 1 keV. In this experiment, electron-beam energies of 700 eV to 1600 eV were used to produce the desired charge-state  $\text{Cl}^{12+}$  for the optical observations. Energies of 10 keV were used for the production of H-like and bare ions, so that the ion loss rate from the stored-ion cloud might be monitored in the x-ray range [22].

The actual ion trap region was imaged by two  $f/4$  10-cm-diameter quartz lenses onto the photodetector, a low-dark rate, half-inch diameter, end-on-cathode photomultiplier (Hamamatsu Type R2557 with a 401 K spectral sensitivity curve).  $M1$  transitions usually dominate the optical spectra of electron-beam ion traps [4,21,23], and thus wavelength selection by interference filter was deemed sufficient, even as several ion charge states were present in EBIT. Two filters were used alternatively; one with a transmission curve centered at a wavelength of 570 nm (bandpass 50 nm, peak transmission of about 50%), the other at 577 nm (bandpass 10 nm, peak transmission of about 70%). Most of the stray light that passed through the filters originated from the hot filament of the electron gun and was further reduced by running the latter at lower than usual electrical currents.

Chlorine was ballistically injected into the trap from a reservoir of trichloroethylene. The gas injector was run at pressures in the range from  $1 \times 10^{-8}$  Torr to  $4 \times 10^{-7}$  Torr. The actual presence of chlorine in the ion trap region was ascertained by producing  $K$  x rays in He-like chlorine ions with an electron-beam energy set at the  $KLL$  dielectronic resonance near 2100 eV. The vacuum pressure readings in the electron gun region below the actual trap were about  $7 \times 10^{-10}$  Torr. The actual vacuum in the trap region is much better, because the surfaces are at liquid He temperature (while the aforementioned vacuum gauge is in a compartment with the hot electron gun). However, there is no direct measurement of the pressure inside the trap volume, and only the pressure range, of normally well below  $10^{-11}$  Torr, can be inferred. This is why any such lifetime measurement needs to be accompanied by a measurement of the ion loss rate (the inverse of the storage time constant) which relates to the vacuum conditions inside the trap region (see below).

For the lifetime measurements, the electron-beam ion trap was operated in a cyclic mode. The data were sorted into 0.1-ms-wide time bins and accumulated over 200–400 min per parameter setting. About every 0.15–0.2 s the accumulated ion cloud was purged from the trap. The electron-beam was switched on for intervals of 30–60 ms, ionizing and

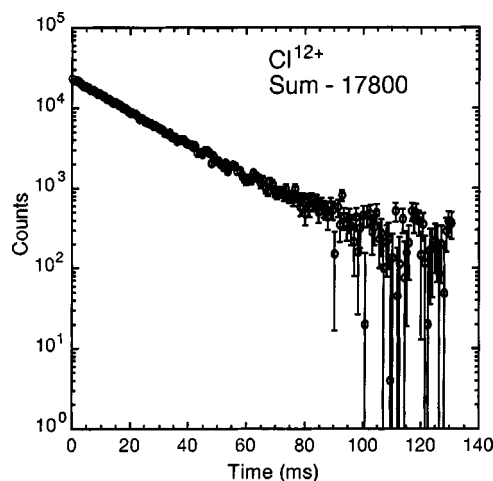


FIG. 2. Photon signal (logarithmic scale) obtained with  $\text{Cl XIII}$ , after the electron-beam in EBIT-I is switched off (magnetic trapping mode). A background contribution of 17 800 counts per channel has been subtracted from the data, leaving about 200 background counts per channel for display reasons.

exciting the ion cloud in the trap. Then the electron-beam was switched off for about 120–150 ms, which let the ion cloud expand to a new equilibrium at a somewhat larger diameter, but still held the ions in the so-called magnetic trapping mode [24]. The switching time of the electron-beam acceleration voltage (“anode voltage”) was of order  $30 \mu\text{s}$ . The electron-beam energies were chosen about 100 to 900 eV above the production threshold of the  $\text{Cl}^{12+}$  ion (i.e., the ionization limit of the  $\text{Cl}^{11+}$  ion) of 592 eV.

The signal rate in the decay part of the datasets ranged up to 900 counts/min. The true dark rate was about 2 counts/s, or 120 counts/min. A total of nine decay curves was collected. The excitation efficiency varies with the electron-beam energy in relation to the production threshold, as does the charge-state distribution in the trap. The statistically best results were obtained at the lowest gas injection pressures and the lowest electron-beam energies. The quality of the various datasets is, of course, reflected in the statistical errors obtained from their analyses. The sum of the data obtained at the lower injection pressures in our range are shown in Fig. 2. In the decay curve analysis, the background was treated as flat. The straightness of the data in Fig. 2 (after subtracting most of the background) corroborates this assumption. Data evaluation was restricted to the part of the decay curves after the first few milliseconds after switching off the electron-beam, in order to avoid possible stray influences of the switching processes or of ion cloud relaxation on the decay curves. The statistical reliability of the EBIT data was sufficient to yield an error bar as little as 1.5%, on a mean lifetime of about 20.5 ms.

The apparent value for the intensity decay rate is modified from the true (atomic) value by the underlying loss rate of ions from the observation zone (by thermal evaporation of the ion cloud along the magnetic field, charge-changing collisions with the neutral rest gas, or diffusion across the magnetic field). The loss rate needs to be established in order to derive the atomic lifetime from the apparent lifetime. Tem-



poral developments in several charge states of multiply charged ions are best studied in the x-ray range, where spectral lines from several charge states can be detected simultaneously with an energy-dispersive detector. However, owing to the filled  $K$  shell, the B-like ions of chlorine do not feature any x-ray transitions. In order to obtain nevertheless an estimate of the ion loss rates, we resorted to charge-exchange (CX) measurements on more highly charged ions that have x-ray decay channels. At electron-beam energies of about 10 kV, plenty of bare, H- and He-like ions of chlorine were produced.

The x-ray detector, a germanium (EG&G Ortec IGLET) detector, viewed practically the same volume as the optical detector. The x-ray spectrum shows light from the production of He- and H-like ions, with H-like ions being apparently less abundant by at least one order of magnitude. Most of this light is from levels that decay promptly, and most of the emission ends when the electron-beam is shut off. However, there is some delayed emission from CX reactions, when bare ions in a collision with neutral rest gas particles capture an electron to become H-like, or when H-like ions capture an electron and thus turn into a He-like ion. It is imperative to do the CX measurement on ions of the same temperature as in the optical experiment, because the rate coefficients depend on the ion velocity. Therefore the same ion trap depth and electron density (beam current) during production have to be maintained. However, producing the more highly charged ions by stepwise ionization takes more time, so the conditions are not quite the same.

The decay curve of the  $K$  x-ray signal showed a time constant of about 240 ms. This ion lifetime, after a linear adjustment for charge state (CX is proportional to the ion charge, at least for fast ions—but we have relatively slow ions), would indicate an ion storage time of B-like ions of Cl of about 320 ms. This, however, is still an underestimate, since experience shows that the capture cross section into the  $K$  shell is much larger than that for capture into the  $L$  shell. In a comparable experiment on  $\text{Fe}^{13+}$ , close in time to the Cl experiment, a storage time of 5 s was found. In that experiment, charge-exchange produced  $L$ -shell x-rays were studied in place of the  $M$ -shell ions of interest. The latter value, adopted to Cl XIII, would require a systematic correction of the measured lifetime by 0.1 ms. A fit to the lifetime data as a function of injection pressure ( $1 \times 10^{-8}$  Torr to  $4 \times 10^{-7}$  Torr) indicates a shift of the mean lifetime by up to 0.2 ms. An ion storage time as short as 500 ms (an estimate derived from the above, assuming a moderate effect of  $K$ -versus  $L$ -shell CX) would necessitate a correction by almost 1 ms. The present experiment yields no clue to resolve this dilemma. We therefore apply a systematic shift by the mean of the alternative corrections, 0.5 ms, and associate with it an uncertainty of the same size. This ion storage time correction uncertainty of almost 2.5% dominates the error budget. Combining all errors, we derive a lifetime value of  $21.0 \pm 0.5$  ms (transition rate  $47.6 \pm 1.1 \text{ s}^{-1}$ ) for  $\text{Cl}^{12+}$  from the Livermore EBIT-I experiment.

### III. DISCUSSION AND CONCLUSION

The general interest in the transitions studied here is reflected in the number of calculations, including very recent

TABLE I. Comparison of predicted and measured lifetimes  $\tau$  for the  $2s^2 2p^2 P_{3/2}^o$  upper level of the ground state in Cl XIII. The computations are labeled by the usual acronyms of the computer algorithms.

$\tau$ (ms)	Reference
Theory	
21.05	[26] Scaled-Thomas-Fermi
20.55	[27] MCDF
21.09	[27] MCDF
21.06	[28] MCHF
21.14	[29] MCDF
21.19	[30] MCDF
21.08	[31] SUPERSTRUCTURE
21.13	[32] RQDO
Experiment	
$(21.2 \pm 0.6)$	Heavy-ion storage ring, this work
$(21.1 \pm 0.5)$	LLNL EBIT-I, this work

<sup>a</sup>Theory results afterwards adjusted for the experimental transition energy.

ones. In Table I, we compare our results only with calculations that explicitly provide transition rates for chlorine. The present lifetime results for Cl XIII, from two very different experiments, agree fully with each other. They are compatible with the theoretical expectations after these are corrected for experimental transition energies, and they also fit to the same isoelectronic trend as do the results of heavy-ion storage ring work [6] and data from the EBIT-II trap on other elements (see Fig. 3). Theory is corroborated at the 2% level of our experiments for the ground complex in Cl XIII, similarly to the heavier ions (with their shorter level lifetimes and hence smaller systematic error corrections), using both the LLNL electron-beam ion trap and a heavy-ion storage ring. Evidently, the usual theoretical uncertainty estimates for these transitions of 10–20% are rather conservative.

At the heavy-ion storage ring, the largest factor limiting

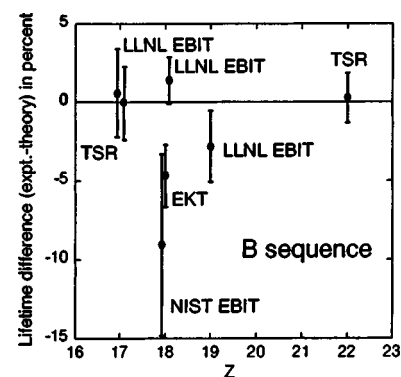


FIG. 3. Lifetime data for the  $2s^2 2p^2 P_{3/2}^o$  level in the ground state of B-like ions from  $\text{Cl}^{12+}$  through  $\text{Ti}^{17+}$ . The data are normalized to the theoretical results given by Galavís *et al.* [31], which include a semiempirical correction for experimental transition energies. The experimental data other than those for Cl XIII are from an electrostatic (Kingdon) trap (EKT) [2], from the NIST and LLNL EBITs [3–5,25], and from the heavy-ion storage ring TSR [6].

the precision of the measurement was the dark rate of the available photomultiplier. At the Livermore EBIT, a PMT with a smaller cathode area and selected for a lower dark rate was available. Here the limiting factor was the precision with which the ion storage time could be obtained. The ion storage time was expected by Wargelin *et al.* to be the limiting factor for any atomic lifetime measurement using electron-beam ion traps [7]. A lower limit of the ion storage time was then estimated to be 100 ms. We now confirm the expectation from a subsequent study [24] that ion storage times in the electron-beam ion trap can be much longer, thus enabling the measurement of atomic lifetimes that might be close to 100 ms, or even longer, as long as the ion storage time can be reliably determined. An insufficient correction for the finite effective ion storage time inside the trap results in an assumed lifetime value that falls short of the true atomic lifetime. Such a deviation, in fact, is evident for several of the earlier forbidden-line lifetime measurements on highly charged ions of argon that were carried out on the NIST EBIT and on an electrostatic (Kingdon) ion trap (Fig. 3).

The present combination of a measurement of the same atomic lifetime in a heavy-ion storage ring (where the systematic errors are smallest) and an electron-beam ion trap (where the choice of elements and ease of operation is much greater) has yielded several benefits. First, the lifetime of interest has been determined with accuracy and the previous

data range on this transition has been extended. This provides a reliable check of theoretical predictions both for a number of individual elements and for an expanded section of the isoelectronic sequence. Second, the present performance limit of the electron-beam ion trap techniques for atomic lifetime measurements in the visible has been tested and established for lifetimes up to about 20 ms. This implies a test for ion traps that have been set up for atomic lifetime measurements: If the present lifetime result on  $\text{Cl XIII}$  or the equivalent lifetimes in  $\text{Ar X}$  and  $\text{Ar XIV}$  cannot be reproduced, the experimental technique must be considered insufficient and the systematic uncertainties higher than presumed. Among all the traps for highly charged ions, so far only the heavy-ion storage ring and the electron-beam ion traps at Oxford [25] and Livermore have passed this test.

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